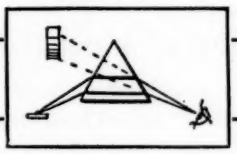
Journal of the

OPTICAL SOCIETY *of* AMERICA

VOLUME 24
No. 12



DECEMBER
1934

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Published monthly
at Prince and Lemon Sts., Lancaster, Pa.
by the
AMERICAN INSTITUTE OF PHYSICS
Incorporated
11 East 38th Street, New York, N. Y.
FOR AND UNDER THE EDITORIAL SUPERVISION OF
THE OPTICAL SOCIETY OF AMERICA

Journal of the
OPTICAL SOCIETY *of* AMERICA

VOLUME 24 DECEMBER, 1934 NUMBER 12

Interference Colors in Oil Films on Water

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(Received October 12, 1934)

CERTAIN types of oil have the property of spreading on a water surface to form films which are sufficiently thin to show interference colors. Recent experiments have led to a method for producing films which exhibit uniform, brilliant color over a very large area. The thickness of the film can be easily controlled by the experimenter, the range of possible thicknesses extending from a small fraction of a wave-length of visible light to more than a millimeter. The thickness is measured directly by placing a known volume of oil on a water surface and measuring the area to which it spreads. Any uniform thickness can be maintained constant for several minutes, or can be increased or decreased continuously by any desired amount. Thus the colors corresponding to several orders of the spectrum of white light can be viewed in succession.

In order to obtain satisfactory results with oil films on water it is essential that the water surface be absolutely clean before the oil is placed on it. Water contained in ordinary receptacles almost always has at least a monomolecular film of grease on the surface. Therefore it is necessary to adopt a technique such as that developed by Langmuir[1] and by Adam[2] for cleaning surfaces of water. The trough devised by Adam is very convenient for optical experiments. It is a brass trough of rectangular shape about 2 cm deep. The sides are at least 1/8 inch thick and are made with their upper edges level so that the trough may be completely filled with water. Flat strips of brass, laid across the top of the trough with their under-surfaces touching the water, serve as movable barriers which are used to sweep the surface clean of grease. The edges of the tray and the barriers are each coated with a thin covering of paraffin wax which keeps them from being wet by water; this makes it impossible for a surface film to leak from one side of a barrier to the other side.

An oil that is well suited to optical experiments may be prepared by oxidizing a lubricating oil, such as a high grade motor oil. Fresh motor oil commonly does not tend to spread on water until it has been exposed to the air for a considerable length of time. If a drop of fresh oil be placed on a clean water surface, the drop will assume the form of a lens having a thickness many times too great to show interference colors. On the contrary, old oil which has been long exposed to the air, such as the oil drained from the crankcase of an automobile, will spread on water to form exceedingly thin films. This occurs because the oil has become partially oxidized, and the oxidized molecules have the property of lowering the surface tension of water, with the result that the oil spreads to a thin film. The more an oil is oxidized, the thinner the film becomes.

[1] I. Langmuir, J. Am. Chem. Soc. **39**, 1848 (1917).
[2] N. K. Adam, Proc. Roy Soc. (London) **A99**, 336 (1921).

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New oil, oxidized in the laboratory, gives more uniform films than used oil, being free from grit. Many types of oxidized oils form thin films on water, but a large proportion are not useful for optical experiments because the films are not stable. For example a pure mineral oil, oxidized, gives a film which commences to develop holes after one or two minutes, the holes gradually becoming larger as the oil draws together into the form of a flat network which ultimately breaks up into individual lenses. Motor oil has given the most satisfactory results of any oil that has been tested. A supply of this oil can be oxidized in the laboratory by heat or by exposure to visible or ultraviolet light. Tests have shown that the product formed by heating the oil gives more permanent films than that formed by exposure to light. The oil should be heated in an open dish and will become rapidly oxidized if kept so hot that it "smokes," and if oxygen is continually supplied by stirring or by a stream of bubbles of air or oxygen.

The oil may be tested at intervals during the oxidation process so that heating may be discontinued when the oxidation has reached any desired stage. In order to test the oil, clean the surface of the water in the trough by sliding a barrier along the trough, and then place a droplet of oxidized oil on the water. After a few minutes' heating at high temperature, motor oil will spread to a film that shows interference colors. With further oxidation the resulting film is colorless but plainly visible; and when oxidation has proceeded very far the film becomes invisible. Use only a tiny droplet so that the oil may spread freely without covering the entire surface of the trough, for if the spreading is at all restricted the film-thickness will no longer measure the degree of oxidation. Small amounts of oil are conveniently transferred by dipping a clean platinum wire into the oil and then touching the wire to the water surface. Clean the wire by heating it to red-heat in a Bunsen flame, then let the wire cool before it touches the oil for a very hot wire will oxidize the oil with which it comes into contact.

Oil which has just reached the stage of giving colorless films is the most convenient for many purposes. I shall call this "oxidized" oil. Suppose, for example, that one wishes to prepare a film that will appear blue, due to the absence of yellow light of the first order spectrum. This may be prepared from oxidized oil by either of two methods (a) or (b).

(a) *Films not confined by barriers.* Dilute the oxidized oil by adding successive small amounts of a non-spreading oil such as a pure mineral oil or an unoxidized motor oil. Test a drop at each stage, giving each drop plenty of space to expand freely on the water. As the oil is made more dilute, the test films, originally colorless, will become thicker and will therefore commence to show color, the first films appearing yellow, the next in succession red, the next blue. Thin films of motor oil are quite rapidly oxidized by the action of light, and a film in bright sunlight can be seen to change slightly in color in about a minute, the color shifting toward shorter wavelengths in the spectrum. Thus a film which originally appears red, due to the absence of green light from the reflected beam, is seen to change to yellow as the color which is absent shifts from green to blue. Changes in film thickness due to progressive oxidation can be obviated by using method (b).

(b) *Films confined by barriers.* This method consists in altering the thickness of a film by compressing it in a confined area. Set one of the barriers at a position say half-way along the trough, and place sufficient oil on the water to fill completely the area between the barrier and the end of the tray. Then move the barrier so as to decrease the area, and the oil film promptly thickens, the change in thickness being made visible by a shifting to successive colors in the spectrum. The change is seen to pass across the film in a wave of changing color, since the film thickens first at the edge adjacent to the moving barrier; as soon as the barrier is halted, the thickness once more becomes uniform. The colors of the film may be made to pass through several orders of the spectrum by this method.

When a drop of oil is placed on a surface which already contains a confined film, the oil is seen to spread outward in concentric rings of vivid color. This effect is most striking when the spreading is made to take place slowly by using an oil of very high viscosity.

The thickness of any uniform film can be determined very accurately by placing on the

TABLE I. *Thickness of oil films on water.*

Vol. (cc)	Area (cm^2)	Thickness (cm)	Color of film	Order of spectrum
0.0173	1365	1.27×10^{-5}	yellow	1
.0284	1686	1.68×10^{-5}	yellow + red	1 1
.0208	861	2.42×10^{-5}	blue	1
.0350	1135	3.08×10^{-5}	green + yellow	1 2
.0505	838	6.03×10^{-5}	red	3

water a known volume of oil and measuring the area which it occupies. In making these measurements it is desirable to compress the oil by at least 10 percent of its area in order to force the oil well up into the corners of the trough so that the area of the film shall be accurately rectangular. For example, if one wishes to measure a red film it is well to use an oil a little more concentrated than the oil which gives a red film when allowed to spread freely, i.e., use an oil which gives a yellow film when not confined, but do not let it spread beyond the area at which the film is red.

Table I illustrates the results obtained by this method of measuring film-thickness. The trough that was used had the dimensions 77×46 cm. Pennzoil Motor Oil was used, which was oxidized at 260°C. The colors were viewed at an angle of incidence of approximately 45 degrees. These data were taken in the course of experiments which were not primarily concerned with the optical properties of the oil and for that reason the index of refraction was not determined.

Photophysical Changes in Silver-Silver Chloride Systems

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(Received August 21, 1934)

In the field of photographic research there appear many puzzling phenomena which in themselves are of interest only as curiosities except insofar as the information which they may yield upon careful investigation leads to a more complete and precise understanding of the fundamental photographic reactions involved in the formation of a latent photographic image and its subsequent development. The action of red polarized light upon certain non-optically sensitized photographic emulsions produces an image which is characterized by polarization and is, in certain cases, photo-adapted so that it appears to be the color of the light by which it was produced. An investigation of this phenomenon in its simplest form was undertaken by the authors in an effort to relate it if possible to accepted photographic theory and to arrive at an explanation which would be more satisfactory than those previously proposed.

Part I: The Effects and Their Origin

AS early as 1870 Seebeck discovered that under certain conditions silver chloride could be affected by light in such a manner that the color of the image was similar to that of the light which produced it. This phenomenon is termed photo-adaptation. Becquerel, using a photo-chloride emulsion, was able to obtain a crude representation of the solar spectrum. It was not until 1919 that Weigert discovered that not only was the color of the image adapted to that of the incident light but that if the light was plane polarized the resulting image was dichroic,[1] that is to say, exhibited variations in color and intensity of transmission when viewed in white light through a Nicol prism which could be rotated. The image was of such a nature that it was more transparent to light of wave-length and plane of polarization identical with the light which produced it than to light of other characteristics. This was termed positive dichroism in keeping with customary photographic nomenclature.†

Weigert found[2] that if shorter exposures than were required to produce a printed-out image were given to suitable photographic emulsions the image rendered visible by suitable development was dichroic but that the sign of the dichroism was negative. This is in contrast with the positive sign of dichroism of the primary effect.

A great deal of attention has been devoted to the developed effect in the past few years and Weigert and his students have examined the influence of variations in exposure and development conditions very minutely.[3] The lack of complete understanding of the development process makes a study of the fundamentals of the effect more difficult and for this reason the authors limited themselves to an examination of the primary effect where the only factors involved were the physical ones of exposure conditions and the chemical ones of the nature of the light sensitive system. Since the uncertainties of development were eliminated it was easier to obtain consistent and reproducible results.

In order to explain the phenomena which he discovered, Weigert postulated a theory of the photographic effect which supposed the existence of a micellar structure in the photosensitive emulsion. The micelles were presumed to consist of gelatin, crystalline and noncrystalline silver halide, photo-silver, "Ursilber," silver sulfide and indeed all the list of compounds which are found in photographic emulsions. These micelles were postulated to undergo a change analogous to a loosening of their structure in planes parallel

* National Research Council Fellow in Chemistry, 1932–33.

[1] F. Weigert, Verh. dtsch. physik. Ges. **21**, 479 (1919).

† The authors will adhere to this sign convention throughout this paper, although Weigert has reversed the sign in his recent publications. Cf. Weigert, Zeits. f. physik. Chemie **B18**, 73 (1932).

[2] F. Weigert, Zeits. f. physik. Chemie **B3**, 389 (1929).

[3] For a summary and bibliography of Weigert's publications dealing with photo-anisotropy the reader is referred to: F. Weigert and J. Matulis, Kollch. Beihefte **38**, 384–412 (1933).

to the electric vector of the incident light. It is not clear how this is supposed to occur nor is it clear why the loosening of the structure in a definite direction should result in the formation of a dichroic image. The loosened structure had to be assumed to be more transparent to light of the correct frequency and plane of polarization but how this could be brought about is not obvious.

The work which is being reported upon here was undertaken in the hope that not only would more facts be discovered concerning the phenomenon of photo-dichroism but that a more satisfactory theory of the effect might be formulated after a study of these facts.

Experimental Method

The photographic emulsions recommended by Weigert are all very fine grained and quite transparent. The silver halide crystals are so small as to be at the limit of microscopic resolution even with an oil immersion lens. The authors found that, apart from specially prepared emulsions, the primary photo-dichroic effect could be obtained with ease on Eastman Kodak "Minimum Grain" spectroscopic plates, Ilford Gaslight Lantern and Imperial Gaslight Lantern emulsions. Of these three the Eastman Kodak plates were the most transparent with the Ilford second.

It may be mentioned here that the developed effect was obtained in bromo-iodide and chloride emulsions specially prepared by Dr. Lambert of the Eastman Kodak Research Laboratory. The former was almost perfectly clear while the chloride showed a slight opalescence. The *p*-phenylenediamine-sodium sulfite physical developer recommended by Weigert[2] was used. The images on bromo-iodide plates appeared a deep blue and were dichroic when viewed by light reflected from the plate at a glancing angle and examined with a Nicol prism set to extinguish light polarized by the plate itself. They were intensely dichroic when examined by transmitted light and showed values of the angle 2α as high as 17° at 6600A with correspondingly intense double refraction.

The exposures were made by illuminating an aperture 16 mm square with the light focused from a five ampere carbon arc and imaging the aperture on the plate with a photographic objective. Filters were inserted in the beam to limit the illumination to a definite spectral region. A Wollaston double image calcite prism was adjusted in the path of the light so that two images of the aperture were formed on the plate in accurate contact along one edge. The photographic image so obtained was its own photometric field since the dividing line between the two areas, which were illuminated by light polarized by the Wollaston prism in two directions at right angles to each other, was reasonably sharp. The image could be examined for dichroism under a simple polariscope or the dichroism and double refraction could be measured by the modified polarimeter described in the second part of this paper.[4]

Exposure of fresh silver chloride emulsions to red light gave no visible print-out image for exposures less than 30 minutes with the apparatus described above and the images then produced were not found to exhibit measurable dichroic effects. Exposure of a silver chloride emulsion to visible or ultraviolet light until photo-silver has been formed produces an emulsion which is red sensitive. This is analogous to the well-known Becquerel effect.[5] The plates used in this work were prepared by exposure to the unshielded light from a 2.5 ampere quartz mercury laboratory arc at a distance of 25 cm for 30 minutes. After such treatment the plates were blue by transmitted light and brownish by reflected light due to the presence of a comparatively large amount of metallic silver produced by photochemical decomposition of the halide. Later in this paper analyses of these plates are given showing the actual quantity of silver present.

Exposure of one of these photo-halide plates to red light produced a visible red image in from 15 to 30 seconds and images visible by polarized light were produced with exposures shorter than this. Maximum intensity of the effect was obtained with exposures of about two minutes. The dichroism and double refraction were strong. With light of shorter wave-length the color-

[4] F. Weigert, Zeits. f. physik. Chemie **B18**, 73 (1932).

[5] Ed. Becquerel, *La Lumière*, Paris, Didot Fréres, Fils et Cie., p. 176, 1867.

adaptation was not pronounced and the dichroism weak.

The fact that red light was so effective in producing adaptation to color and polarization suggested that the phenomenon was akin to the Herschel effect, i.e., destruction of the latent image by infrared radiation. It is known that a great many organic dyes are light sensitive when used to stain gelatin layers and that photo-dichroism can be produced in such systems.[6] It was imagined that perhaps in some similar fashion the photo-halide was selectively "bleached" by the polarized red light. Consequently an attempt was made to obtain a photo-dichroic image with infrared radiation. The carbon arc was used with an infrared transmitting filter of a saturated solution of iodine in carbon disulfide. An 88-A Wratten gelatin filter was also tried. With both filters an aqueous solution of potassium chromate was employed as well in order to ensure the absence of ultraviolet and long wave-length heat radiation. In neither case could more than a trace of image be found even after exposures of as much as four hours duration. The idea of photo-dichroism as a kind of selective Herschel effect was therefore abandoned.

It was found by the present authors that the primary photo-dichroic effect was produced most easily and consistently with red light and finally all the exposures were made with the light from a five ampere carbon arc filtered by a Wratten red filter No. 25 (Standard Tricolor Red) and a 7.5 cm cell of a strong aqueous solution of potassium chromate.

The Origin of the Effect

The first part of the work consisted in a search for the necessary and sufficient conditions for the production of the primary effect—i.e., the production of a color-adapted image upon exposure to red light without subsequent development.

(a) Silver chloride is necessary

The red sensitization of silver halide emulsions by photo-silver is well known in the Becquerel effect which has already been given mention. It was thought necessary to establish the fact that silver alone was incapable of showing photo-adaptation. Emulsions of colloidal silver in gelatin were coated on glass and given varying exposures to light of several spectral regions but under no conditions of illumination could any visible image be obtained.

(b) Silver is necessary

Unexposed plates of various types were taken from their wrappings in the dark and exposed to red light from the arc lamp for times varying from 30 seconds to 30 minutes and in no case was any adapted image produced. The same plates would give a red dichroic image if they had been previously exposed to blue or ultraviolet light until the plates themselves were blue. According to Hilsch and Pohl[7] the blue color is due to the induced absorption produced by particles of metallic silver within the crystal lattice. This is quite different from the coloration of alkali halides by photo-liberation of electrons from the halogen ion for in such cases the coloration is produced by the alkali atoms themselves and not by aggregates of atoms.

(c) The silver can be deposited chemically

The necessary conditions for the formation of the color-adapted image having been proved to be the coexistence of both silver and silver chloride, the effect of depositing the silver chemically instead of photochemically was examined. For this purpose an Ilford Gaslight Lantern plate was soaked in dilute ferrous oxalate developer* (or in Eastman Kodak D-72 diluted to ten times normal concentration) for five minutes in the dark and briefly rinsed. It was then exposed to illumination from tungsten lights in the dark-room until fogging became apparent. The plate was then washed for an hour and a half in darkness and dried. The plates

[6] T. Kondo, Zeits. f. wiss. Photogr., Photophysik, Photochemie **31**, 153 (1932).

[7] Hilsch and Pohl, Zeits. f. Physik **77**, 421 (1932).

* Solution *A*: $K_2C_2O_4.H_2O$ 142 g; Water (hot) 568 ml
Solution *B*: Water (warm) 568 ml; H_2SO_4 (Conc.) 1.8 ml; $FeSO_4.7H_2O$ 142 g
30 ml *A* + 10 ml *B* + 200 ml water

so produced were reddish brown when viewed by transmitted light. Upon exposure to red polarized light they became photo-adapted and polarimetric examination showed that the dichroism was strong. The silver in this case was deposited chemically on the surfaces and edges of the silver halide crystals and was not present as photo-silver within the crystal lattice.

(d) Effect of " packing " of the emulsion grains

The fact that only transparent emulsions show the photo-dichroic effect in any marked degree is due merely to the scattering and depolarization of the incident light which is produced in a coarse grained emulsion. The absence of any marked photo-dichroism in coarse grained emulsions has, however, led to misconceptions for it might appear that the effect was connected in some manner with the passage of photoelectrons from one grain to its neighbor. Coton[8] tested this by examining the effect of a magnetic field of 50,000 gauss parallel to the direction of the incident light but obtained a negative result.

To see if closeness of packing of the silver halide grains had any influence the emulsion scraped from a soaked strip of plate sensitized by exposure to ultraviolet light was diluted with 20 times its own weight of soaked gelatin and recoated. The plates exhibited as good photo-adaptation as before. Some of this emulsion was diluted with water to 4000 times the original bulk of the dry film and allowed to set to a gel in a glass cell. The gel exhibited photo-adaptation to color and direction of polarization.

(e) Photo-adaptation in sols

To determine whether the gel structure exerted any influence upon the effect a colloidal suspension of silver chloride was prepared by precipitating extremely dilute silver nitrate with correspondingly dilute sodium chloride in the presence of a trace of gelatin to stabilize the sol. Care was exercised to ensure a slight excess of chloride ion. After exposure to ultraviolet light the sol turned blue because of the formation of photo-chloride. The blue sol turned red after an exposure of one or two minutes to high intensity red light. The gel structure therefore plays no essential part in the photo-adaptation mechanism.

(f) Photo-adaptation in the absence of gelatin

The first attempts to produce a blue sol of photo-silver in absence of gelatin failed. If, however, the precipitation of the silver chloride from pure dilute aqueous silver nitrate and sodium chloride was allowed to take place in the illumination from a quartz mercury arc sufficient photo-silver was formed to impart a blue color to the sol. This suspension was adequately red-sensitive and turned red after a short exposure to red light. This satisfied the hypothesis that the photo-adaptation occurs in a silver-silver chloride system and that all other components of the emulsion are superfluous. This disposes of theories which presuppose the existence of micelles containing gelatin.

(g) Photo-adaptation in single crystals of silver chloride

To confirm the idea that was stated in the last paragraph an attempt was made to produce the effect in single crystals of silver chloride. The crystals were grown from an ammoniacal solution of silver chloride by slow evaporation of the ammonia. The silver chloride was prepared from pure solutions of silver nitrate and sodium chloride with an excess of chloride ion and was precipitated, thoroughly washed and dissolved in ammonium hydroxide before exposure to actinic light.

Several days were required to grow crystals about 0.5 mm on an edge. If these crystals were exposed to blue or ultraviolet light they turned blue owing to the formation of photo-silver. This occurred best if the mother liquor had contained a trace of gelatin. It appeared that traces of some component of the gelatin might be occluded in the lattice during crystal growth and served as halogen acceptors. It was then found that blue crystals of silver chloride could be produced by illuminating the solution with a quartz mercury laboratory arc during the evaporation of the ammonia.

The blue crystals prepared in this manner, as well as those grown from a solution containing a trace of gelatin, showed both color-adaptation and dichroism after exposure to red polarized

[8] Coton, Comptes rendus **189**, 599 (1929).

light upon the stage of a petrographic microscope. The crystals became dichroic, changing from blue violet to rose pink when rotated on the stage of the microscope in white polarized light and viewed without the analyzer.

This shows unambiguously that the phenomenon of photo-adaptation is one which occurs in a silver-silver chloride system and that all other factors are immaterial. The question of the nature of this change and the mechanism of the effect will be considered in the second and third parts of this paper.

Summary

(1) Photo-anisotropy, both primary and developed, has been produced in several photographic emulsions.

(2) No visible effect was produced by infrared illumination.

(3) Silver and silver chloride together are required to produce the effect. The silver can be introduced chemically as well as photochemically.

(4) Closeness of "packing" of emulsion grains is immaterial.

(5) Gelatin is not required for the production of photo-adaptation and the effects have been produced in single crystals of pure silver chloride.

Part II: Dichroism and Birefringence of the Unfixed and Fixed Photo-Adapted Image

In Part I of this paper the authors have discussed the origin of the phenomenon of photo-adaptation. This paper will deal with the methods used in investigating more closely the mechanism of this phenomenon.

Apparatus

The method of measurement of dichroism and birefringence was that described by Weigert.[9] An Adam Hilger polarimeter, with the specimen trough removed, was modified by the addition of a rotating stage between the polarizer and analyzer. This held the specimens with the optic axes inclined at 45° to the plane of vibration of the light. The source of light was a 250 watt tungsten filament projection lamp and monochromator, the latter being an Adam Hilger constant deviation spectrometer with an exit slit substituted for the eyepiece. The slits were kept fixed and both were set 100A wide at a wave-length of 5850A. The rotating head of the analyzer was fitted with a subsidiary rotating head which could be locked in any desired position relative to the analyzer and which held a Senarmont compensator. This selenite plate of 140 $\mu\mu$ retardation was mounted in a slide with its optic axis set accurately at 45° to the length of the slide so that turning the compensator over side for side in its slot had the effect of rotation through 90° and therefore interchanged the fast and slow axes.

The routine of measurement was described by Weigert[9] and will be reviewed briefly. The photo-adapted image is in two parts and the optic axis of one-half is parallel to the dividing line while that of the other half is perpendicular. The image is adjusted so that the dividing line is a diameter of the field of view at 45° to the direction of vibration of the polarized light. Extinction of any one part of the multiple field can only be obtained if the axes of the compensator coincide with the major and minor axes of the vibration ellipse of the light from that part and if at the same time the analyzing prism is

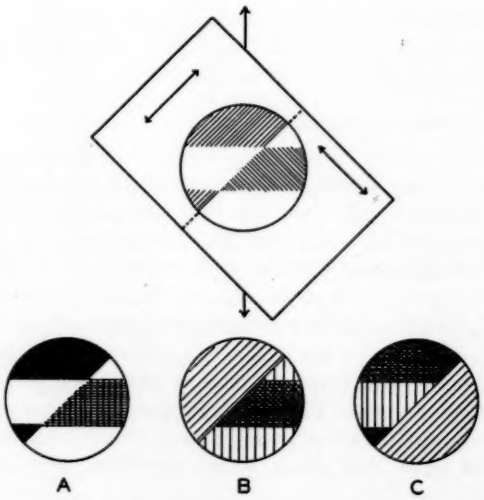

Fig. 1. Appearance of dichrometric fields.

[9] F. Weigert, Zeits. f. physik. Chemie **B18**, 73 (1932).

set so that its principal plane is at right angles to the resulting linear vibration which emerges from the compensator. This double adjustment, Fig. 1A, having been made for one part of the field of view the compensator is locked relative to the analyzer and a match obtained between the appropriate parts of the field by rotation of the analyzer head. All parts of the Lippich prism field seen through one-half of the photo-adapted image are then of uniform intensity, Fig. 1B. The verniers on the polarimeter head are now read to 0.01° of arc.

The compensator is now turned over so as to interchange fast and slow axes and a match obtained for the other half of the field, Fig. 1C. Without changing the compensator, the rotating head is unlocked and extinction again obtained for the part of the field originally chosen. One-half of the field is then matched, the compensator turned over and the other half matched. The tangent of half the angle, 2β, between the first and third and the second and fourth settings of the polarimeter head is the ratio between the major and minor axes of the vibration ellipse.

Because of dichroism the major axes of the vibration ellipses of the light transmitted through the two halves of the photo-adapted image are not parallel. The difference between the first and second readings and the third and fourth gives the angle 2α between the major axes. This is related to the dichroism by the expression: $Di = 2 \log \tan (\pi/4+\alpha)$. The angle 2β is related to the birefringence by the expression $Do = \beta\lambda/180$.

The Senarmont compensator is only accurate at a wave-length for which it produces a retardation of a quarter of a wave but Weigert[9] has shown that for all practical purposes no increase in accuracy is obtained by the use of a Babinet-Soliel compensator and the inaccuracy introduced by the use of a Senarmont compensator over the visible range of the spectrum may therefore be neglected.

Measurements of the Unfixed Image

The curves of dichroism, plotting the more convenient angle 2α instead of $2 \log \tan (\pi/4+\alpha)$, of the red-adapted image are all of characteristic shape. There is always a zero of dichroism and a maximum of birefringence at approximately

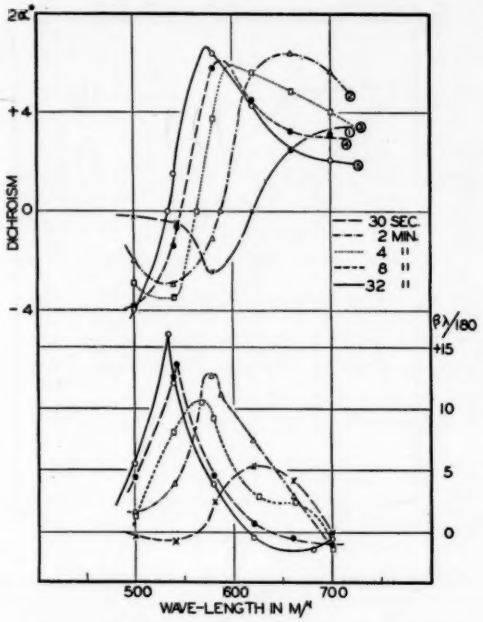

FIG. 2. Dichroism and birefringence of unfixed photochloride plate as a function of exposure time and wave-length of measuring light.

the same wave-length. This point is shifted towards shorter wave-lengths by increase of exposure and reaches a limiting position at about 5350A for long exposures. The curves illustrating these change are shown in Fig. 2.

The dichroism is a result of the difference in absorption of the two components of the incident light. One component vibrates parallel to the original direction of vibration of the light used for exposure and the other at right angles to it. The dichroism should therefore be the difference in absorption of the red photo-adapted image made in unpolarized light and the absorption of the unadapted background.

As a test of this point absorption curves were made by the use of a Bausch and Lomb spectrograph fitted with a multiple Lummer-Brodhun cube in front of the slit and a logarithmic spiral sector rotating in the comparison beam.[10] The photograph of the multiple spectrum was spotted for points of equal photographic density in the

[10] Brian O'Brien, Bull. Am. Phys. Soc., Feb. 10, 1932; J. O. S. A. **22**, 427A (1932).

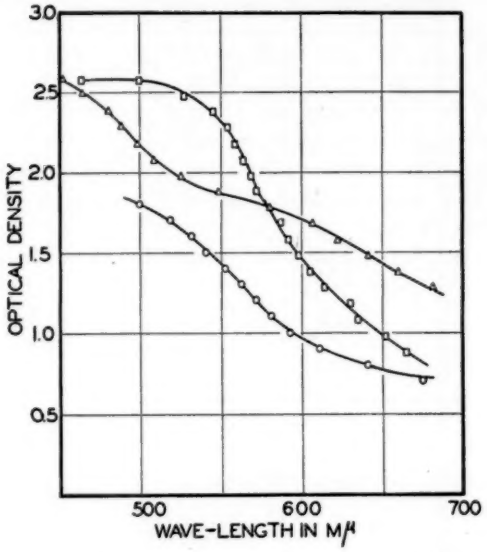

FIG. 3. Circles, absorption curve of unexposed Ilford lantern plate; triangles, absorption after ultraviolet exposure; squares, absorption after exposure to red light.

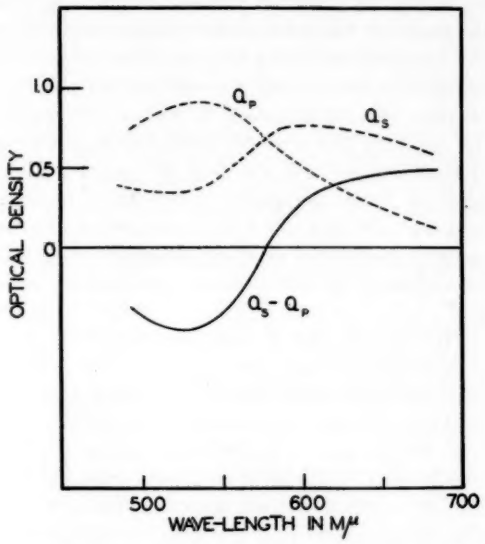

FIG. 4. True absorption of ultraviolet exposed plate, a_S and of red exposed a_p. The difference between these gives the dichroism curve, $a_s - a_p$.

usual manner. The curves obtained for the absorption of the unexposed photographic emulsion, of the same emulsion after exposure to ultraviolet light and after exposure to red light following the ultraviolet exposure are shown in Fig. 3. The corrected curves for absorption by the photo-silver and by the red-adapted image are shown in Fig. 4. The absorption of the unexposed plate is largely due to scattering by the emulsion grains and not to photochemical absorption in the red end of the spectrum.

The difference curve obtained by subtracting one of these absorption curves from the other is exactly the dichroism curve as measured directly. That is to say, the red-adapted image is dichroic simply because light vibrating perpendicular to the plane of vibration of the original exposure is absorbed by the red image according to the same law of absorption as by the unexposed background. It is only the absorption of the component of the light parallel to the original vibration which is altered.

The most remarkable feature of these absorption measurements is the increased opacity of the red-adapted image in the green. That is to say, the process of photo-adaptation is not a mere bleaching process in which the opacity is selectively weakened but is a more complicated phenomenon in which the opacity is simultaneously lessened in the red and increased in the green.

This suggests that upon photo-adaptation there is no change in the total quantity of photo-silver as Weigert has already suggested. Plates were prepared by exposure to ultraviolet light and one-half of the area of each was exposed to red light for a sufficient length of time to give maximum brilliance of red-adaptation. Through the kindness of Dr. S. E. Sheppard of the Eastman Kodak Research Laboratory these areas were then analyzed for free silver. It was found that in the blue and red areas there were present the same amounts of silver within the accuracy of the method. The results of two determinations are given in Table I. Thus the phenomenon of

TABLE I. *Free silver in mg/100 cm².*

Blue	(A) 1.12	(B) 1.12
Red-adapted	1.10	1.12

photo-adaptation does not involve any appreciable change in the quantity of free silver and the origin of the effect must lie in a transformation of the photo-silver, a photoelectric rather than a photochemical effect. The re-

arrangement of the silver in the blue photo-halide as a result of exposure to red light must have the result of diminishing the absorption of red light polarized so that the electric vector has the same direction as that of the original exposure and of increasing its absorption of green light similarly polarized. This point must be kept in mind for it proves to be the key to the explanation of the effect.

Discussion of Measurements

Where does the double refraction of the photo-adapted image originate? Gelatin under strain exhibits double refraction and gelatin films coated on plates are always strained after drying. However, a gelatin film thus prepared has its axis of strain and therefore its optic axis perpendicular to the film. The red-adapted image is doubly refracting with its optic axis in the plane of the film. Moreover, swelling the gelatin in water does not destroy the dichroism or birefringence so that the origin of the double refraction is not to be sought in strain of the gelatin.

It was shown in the first part of this paper that photo-adaptation could be produced in single crystals of silver chloride. Furthermore, the two effects of dichroism and birefringence are so intimately connected, as is seen in the behavior of the curves in Fig. 2, that the explanation of both effects must be sought together.

If a vectorial absorption coefficient is assumed, having components a_s and a_p perpendicular and parallel to the electric vector of the incident light used for the red exposure, then the dichroism of the red-adapted image is the difference between these components. From the evidence of the absorption curves in Figs. 3 and 4 the coefficient a_s is also the absorption coefficient of the photo-silver before adaptation and a_p that of the photo-silver after adaptation. Moreover, if the dichroism is zero and changes sign at any wave-length the curves of spectral variation of a_s and a_p must cross at this wave-length.

The Lorentz dispersion equation connects refractive index with absorption and describes the characteristic behavior of the index in the neighborhood of an absorption band. This is illustrated in Fig. 5. Two dispersion curves, n_s and n_p which result from two absorption curves, a_s and a_p, are shown diagrammatically in Fig. 6 and the difference curve is the birefringence, $n_p - n_s$. It will be seen how closely the experimental curves correspond to the theoretical. It is to be inferred that the dichroism and bire-

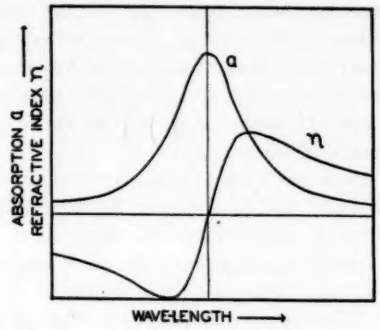

Fig. 5. Typical Lorentz equation for absorption and refractive index. (After Försterling, *Lehrbuch der Optik*, p. 86.)

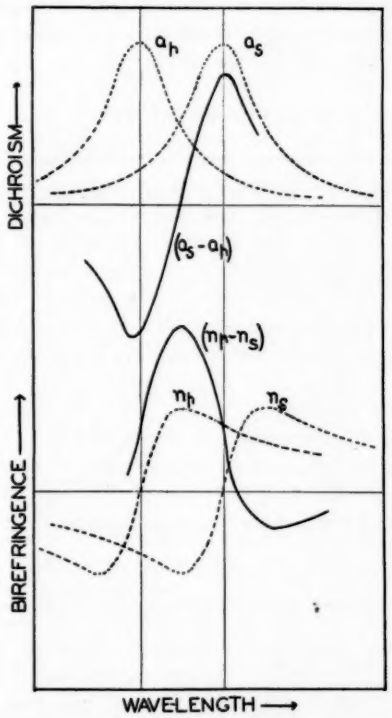

Fig. 6. Two absorption curves (dotted) giving rise to a dichroism curve and two corresponding refractive index curves giving a birefringence curve by difference.

fringence are both functions of the vectorial absorption coefficient of the adapted photo-silver and there is no need to invoke the aid of micellar deformation to explain the effects. The silver and the silver alone is the cause of the dichroism and birefringence.

The question of what causes the two maxima of absorption a_s and a_p is yet to be answered. The blue color of photo-chloride is due not to silver atoms but to metallic particles or aggregates of silver atoms deposited in the silver chloride lattice. What is the nature of the change in the photo-silver upon red-adaptation which alters its maximum of absorption parallel to the electric vector from the red to the green region of the spectrum? Before answering these questions the dichroism and birefringence of the images after fixing will be discussed.

Measurements of the Fixed Image

The red-adapted image on fixing in alkaline hypo or in ammoniacal sodium sulfite solution to avoid dissolving silver is observed to change from bright red to a clear lemon yellow while the background of unadapted photo-silver changes from a blue to a reddish brown. The spectral absorption of these two variations were measured and the curves are shown in Fig. 7. It is seen that the effect of fixing is to have shifted the absorption maxima towards the ultraviolet. The dichroism and double refraction are shown in Fig. 8. These curves are essentially the same as those obtained from measurement of the unfixed image but are displaced toward shorter wave-lengths.

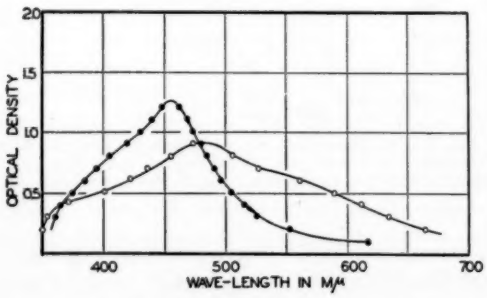

Fig. 7. Absorption curves of plates after fixing. Open circles, ultraviolet exposed plate. Solid circles, absorption of red exposed area.

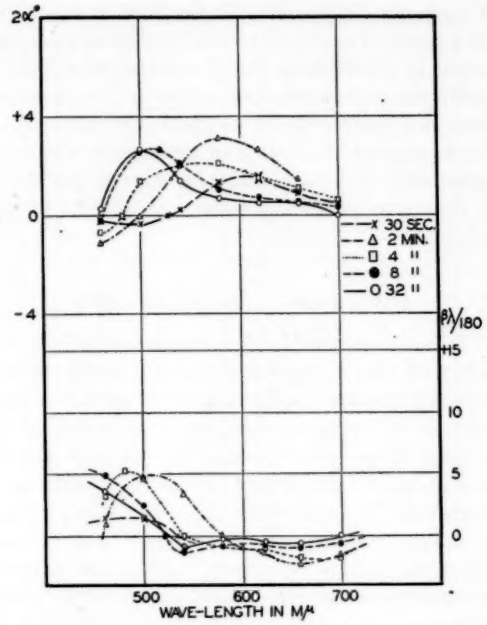

Fig. 8. Dichroism and birefringence of fixed photo-chloride plate as a function of exposure time and wave-length of measuring light.

The shift of the maximum of a_s is from λ6150A to λ4750A or a wave-length ratio of 1.3. The shift of the maximum of a_p is from λ5400A to λ4500A or a wave-length ratio of 1.2. The medium surrounding the silver particles has changed from silver chloride of refractive index 2.06 to gelatin of refractive index 1.53, and the index ratio is 1.3.

The fixed and unfixed images have only one component in common beside the gelatin and that is the silver. These silver particles in the unfixed image are in silver chloride and in gelatin in the fixed image. The dichroism and birefringence as well as the absorption are so similar in the fixed and unfixed image that the silver particles must be held responsible for them in both; a conclusion which had earlier been reached in the case of the unfixed image.

Conducting Particles in an Electromagnetic Field

The classical problem of conducting particles in an electromagnetic field has been studied by

Rayleigh,[11] Abraham,[12] Thomson[13] and Love.[14] The solution of the particular problem of a conducting sphere of radius a in a medium of dielectric constant k is comparatively simple and yields the result that the sphere resonates as a heavily damped Hertzian oscillator to electromagnetic waves of length $4\pi a/3^{\frac{1}{2}}$, that is to say to a wave-length in vacuum of $\lambda=4\pi a(k/3)^{\frac{1}{2}}$. For optical purposes variations in permeability may be neglected and the refractive index $n=k^{\frac{1}{2}}$ introduced so that $\lambda=4\pi an/3^{\frac{1}{2}}$. In other words, if the medium surrounding the oscillator is changed the resonant frequency is changed in the ratio of the refractive indices. This is precisely what is observed to happen to the absorption maximum a_s of the photo-silver when the plate is fixed. To a rough approximation this is also true of the maximum a_p of the adapted photo-silver. It appears likely that the absorption bands of the silver particles in the unfixed and fixed images are due to optical resonance, a phenomenon observed when light is incident on small conducting metal particles.[15, 16]

In verification of this idea, the fixed image was measured for dichroism dry and also while the gelatin was swollen with water. The absorption curves were not measured, but it will be seen from Fig. 9 that there is a further shift of the dichroism curves toward the ultraviolet on reducing the index of refraction of the medium surrounding the silver particles from 1.53 to something approaching 1.35. Unfortunately it is not possible to make a quantitative check from the dichroism curves.

Rayleigh[11] has examined the problem of a conducting ellipsoid of three unequal axes and his equations give the scattering of light by such a particle. If the long axis of the particle is parallel to the electric vector of the incident light the scattering (for long wave-lengths) is greatest but if the period of the wave corresponds to the period of oscillation of the particle the effect will be enhanced by resonance.

[11] Rayleigh, Phil. Mag. **44**, 28 (1897).
[12] Abraham, Weid. Ann. **66**, 435 (1898).
[13] Thomson, *Recent Researches in Electricity and Magnetism.*
[14] Love, Proc. London Math. Soc. **1**, 37 (1903).
[15] Wood, Phil. Mag. **3**, 396; **4**, 425 (1902); **6**, 259 (1903).
[16] Braun, Ann. d. Physik **16**, 1 (1905).

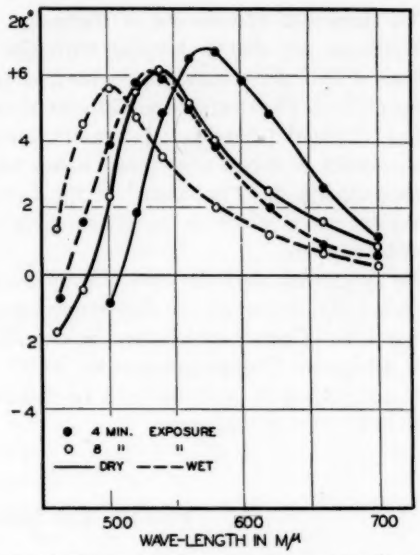

FIG. 9. Dichroism of fixed image wet and dry showing shift of curves toward ultraviolet.

The selective scattering of light by the silver particles may be shown by dissolving the fixed photo-adapted image in water and placing this suspension and a similar one prepared from the background in the path of a powerful beam of white light. The scattering is observed to be markedly different. Both solutions scatter light so strongly as to appear almost fluorescent but as would be expected the lemon yellow suspension of the dichroic image scatters blue and the reddish brown suspension of the background scatters green.

If it be admitted that the color of the image is due to optical resonance, the dichroism must be due to orientation of the particles. These must be arranged with their long axes so that the light which is absorbed by resonance is predominantly red when the electric vector is perpendicular and predominantly green when it is parallel to the direction of the electric vector of the original exposure. After fixing the silver particles are in a medium of lower refractive index but as their orientation is unchanged the image is still dichroic and birefringent. The curves of dichroism are shifted toward the ultraviolet and a further displacement occurs, as is required by theory, when the refractive index is again lowered by soaking the gelatin in water.

The shapes of the curves of dichroism and birefringence are also in keeping with the hypothesis of optical resonance of oriented particles of photo-silver. The electrical oscillations of a conducting ellipsoidal particle are heavily damped which results in broad absorption bands and a correspondingly flat "anomaly" in the Lorentz dispersion curve which is again in agreement with observation.

The hypothesis that the optical properties of the photo-adapted image are due to resonance of the particles of photo-silver may be considered to be adequate. The mechanism by which the photo-adaptation is produced will be discussed in the third part of this paper.

Summary

(1) The apparatus and routine of measurement of the dichroic plates is briefly described and curves reproduced showing the relationship between dichroism, double refraction and wave-length of the measuring light.

(2) Absorption curves of the unexposed plates and of the photo-halide both before and after photo-adaptation are reproduced. Exposure of the photo-chloride to red light causes a decrease in opacity to red light and an increase in opacity to green.

(3) Chemical analysis reveals no change in silver content caused by exposure to red light.

(4) Absorption curves of the fixed plates show a shift toward the ultraviolet of the magnitude expected for conducting particles when the refractive index of the medium is changed from that of silver chloride to that of gelatin.

(5) The optical properties of the photo-adapted image are due to resonance of particles of metallic silver.

Part III: The Mechanism of Photo-Adaptation

In the first part of this paper the process of photo-adaptation was ascribed to a photoelectric effect rather than to a photochemical change in silver-silver chloride systems and in the second part measurements of the effect led to the conclusion that the color, dichroism and double refraction of the photo-adapted image are due to optical resonance of oriented silver particles. In this part of the paper a mechanism by which the effect may be supposed to take place is advanced and experiments to test the theory are described.

The absorption curves shown in Fig. 4, Part II, were obtained from measurements with Ilford Gaslight Lantern Plates—i.e., a gelatin coating containing sub-microscopic and microscopic crystals of silver halide. Curves of absorption by single crystals of silver halides have been obtained by Hilsch and Pohl.[17] The opacity of the photo-silver at any particular wave-length within the induced absorption band is greatly reduced by intense illumination with light of that wave-length in the same way that was found for a photographic emulsion. (Cf. Fig. 4, Part II.)

This must mean that the silver particles which resonate to any particular wave-length are destroyed by prolonged illumination. The production of dichroism by illumination of single crystals of silver chloride by plane polarized red light means that this destruction of the resonators is selective and that only those particles of silver which are suitably oriented with appropriate axes parallel to the electric vector of the illumination are destroyed, since appreciable resonance will occur only if these two conditions are fulfilled. Firstly, the dimensions of the particle must be right and secondly, there must be a large enough component of the electric vector of the incident light parallel to the resonant axis of the particle.

The two observed facts, that the total quantity of free silver remains unchanged in photo-adaptation as shown by chemical analysis and that the opacity is simultaneously increased in the green and decreased in the red, indicate that the resonators are not entirely destroyed but merely diminished in size and increased in number, so that the resonance occurs for shorter wave-lengths. This very marked increase of opacity in the green does not appear in Hilsch and Pohl's curves and it is not clear why this should be so for our measurements are very definite on this point.

The increase of the total number of particles of silver as a result of the redistribution by photo-adaptation is confirmed by the effect of physical development of the fixed images. The unfixed red image on a background of blue photo-halide becomes a lemon yellow image on a background

[17] Hilsch and Pohl, Zeits. f. Physik **64**, 615 (1930).

of reddish brown after fixation with alkaline hypo. A physical developer, such as an alkaline solution of *p*-phenylene diamine and silver nitrate, deposits silver upon existing silver nuclei and if such a developer be used to intensify the fixed image the image is reversed and becomes a dark chocolate brown image upon a lighter reddish brown background. This can be due only to a greater density of silver nuclei in the photo-adapted image than in the unadapted background.

The hypothesis that red illumination in some way redistributes the total silver and forms a larger number of smaller particles which no longer resonate to red light but which are capable of resonating to and absorbing light of some shorter wave-length is confirmed.

Dichroism Produced by Unpolarized Light

The hypothesis of optical resonance demands that a resonator must be of appropriate dimensions in order to absorb light of a definite frequency. If the incident light is plane polarized those resonators which are wrongly oriented cannot respond but those which can respond absorb light and as a result of this response are diminished in size, the excess silver being transferred to some other point in the silver halide lattice. The resulting image is consequently more transparent to light of which the frequency and electric vector are identical with those of the original illumination. In short, it is color-adapted and photo-dichroic. If the original illumination was with unpolarized light the color adaptation is the same but the dichroism is absent. Nevertheless, if the light was plane parallel the electric vector was confined to the plane perpendicular to the beam and consequently the color-adapted image which it produced is not dichroic in the usual manner but should show dichroism if viewed at right angles to the direction of exposure.

In order to verify this a cell was filled with a warm solution of gelatin and water in which was suspended the blue photo-halide from a portion of Ilford plate which had been exposed to ultra-violet light. When the gelatin had set to a gel a parallel beam of unpolarized red light was passed through the cell with the result shown diagrammatically in Fig. 10. The beam left a track of red-adapted photo-chloride which was naturally not dichroic when viewed along the axis of the beam but which was dichroic when viewed in any direction at right angles. The track was more transparent to red light of which the electric vector was perpendicular to the length of the track. In fact, when observed through a Nicol prism the track stood out strongly from the surroundings unless the electric vector of the light reaching the eye was parallel to the axis of the beam, when the track became completely invisible.

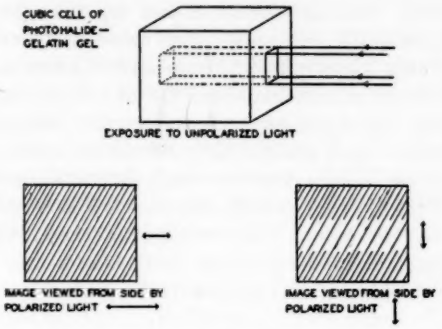
CUBIC CELL OF PHOTOHALIDE—GELATIN GEL
EXPOSURE TO UNPOLARIZED LIGHT
IMAGE VIEWED FROM SIDE BY POLARIZED LIGHT
IMAGE VIEWED FROM SIDE BY POLARIZED LIGHT

Fig. 10.

Résumé of the Pertinent Evidence

It is an almost inevitable conclusion that the color, dichroism and birefringence are due to optical resonance of particles of silver but as to how these are produced of the correct dimensions and orientation there is little indication. There are, however, certain facts which must be remembered which will aid in formulating a mechanism. One of these is that the changes produced by red illumination are purely photo-electric, not photochemical, and that the total quantity of silver remains unaltered. Also, while the dichroism and double refraction are due to silver resonators and persist so long as they persist and remain in their relative orientation even after removal of the silver chloride by fixation, the process of photo-adaptation can go forward only in the presence of the silver halide. Finally the wave-length most effective in photo-adaptation is one of conspicuously low quantum energy,

6500A, whereas the threshold wave-length of photoelectric emission from massive silver in vacuum is about 3300A. Pure silver chloride has a threshold absorption about 4100A and the maximum photographic, photoelectric, photoconductance and photovoltaic effects are produced at wave-lengths generally much shorter than this.

The inferences which may reasonably be made from the totality of observations are these. When light of wave-length less than 4100A falls on silver chloride and is absorbed a photoelectron is ejected from the chlorine ion, leaving behind a chlorine atom. The photoelectron wanders through the crystal lattice and attaches itself to a silver ion, forming a silver atom.[18, 19] If the chlorine atoms can escape or are captured by a halogen acceptor the production of silver atoms can proceed until large aggregations of silver are formed within the lattice. These will be unlikely to be spherical but will have their long axes along planes of weakness in the crystal.[20, 21] The classical electromagnetic theory of light predicts that if these aggregates are of suitable size they will act as damped Hertzian oscillators and resonate to light of a definite range of frequency.

What is observed is that under the influence of intense red illumination the silver aggregates grow smaller but more numerous. This can only be supposed to take place by the transfer of electrons from the neutral aggregates of metallic silver leaving behind ions of silver, the electrons wandering through the crystal lattice until they reform metallic silver at other points by combination with silver ions.[22] Tollert in an investigation of the Herschel effect concluded that red light causes a redistribution of silver by the above mechanism.[23] The quantity of silver present in the plates which he used in the investigations was about 0.2 mg per 100 square centimeters or about one-sixth of the quantity present in the plates used in the investigation of the photo-adaptation effects. (Cf. Part II of this paper.)

The process of redistribution of silver can go forward only if the reverse migration of the chloride ions can take place, for if silver ions are being converted into silver atoms in one region of the lattice and silver ions are being formed from metallic silver in another, chloride ions must migrate to reform the silver chloride lattice in this latter region.

In both the photovoltaic[24] and the photoconductance effects the two processes, ejection of an electron and subsequent migration of the halogen atom, are quite distinct and separate in time. The electron current in the photoconductance effect appears to be more or less independent of temperature but the positive current ceases at low temperatures where migration of the atoms is no longer possible. Photo-adaptation, since it is supposed to proceed in similar stages should be inhibited by low temperatures, because under such conditions the chloride ions could no longer migrate in the lattice and accumulation of space charge would stop the loss of electrons from the silver particles.

The experiment of cooling photographic plates which had been previously exposed to ultraviolet light to produce the blue photo-chloride was tried with liquid nitrogen. At this temperature it was illuminated with red light in the usual manner of producing photo-adaptation but no visible image was produced no matter how long exposures were given. When it is remembered that 30 seconds illumination at room temperature suffices to produce a bright red image it will be seen how complete is the inhibition by low temperatures. It may be noted here that the production of blue photo-chloride by ultraviolet light is similarly inhibited by low temperatures, presumably for the same reason; that the chlorine atoms are frozen in the lattice and cannot migrate. Sheppard, Wightman and Quirk[25] found

[18] S. E. Sheppard and A. P. H. Trivelli, Phot. J. **61**, 403 (1921).

[19] Fajans, Chem. Ztg. **45**, 666 (1921). Zeits. f. Electrochemie **28**, 299 (1932).

[20] Fajans and Frankenburger, Zeits. f. physik. Chemie **105**, 253 (1923).

[21] A. P. H. Trivelli and S. E. Sheppard, Phot. J. **63**, 334 (1923).

[22] S. E. Sheppard, J. Frank. Inst. **210**, 587 (1930).

[23] H. Tollert, Zeits. f. physik. Chemie **140**, 355 (1929).

[24] Vanslow and Sheppard, J. Phys. Chem. **33**, 331 (1929); Sheppard, Vanslow and Hall, ibid. **33**, 1403 (1929); Toy and Harrison, Proc. Roy. Soc. (London) **A127**, 631 (1930).

[25] Sheppard, Wightman and Quirk, J. Phys. Chem. **38**, 817 (1934).

considerable drop in both natural and optical sensitivity of photographic emulsions from −70°C to −180°, with about 5 percent of the original threshold remaining at the latter temperature. They suggest that this may be produced by inhibition of halogen acceptation at this temperature because the chlorine atoms cannot migrate. Hilsch and Pohl[26] found no change in the absorption spectrum of the color centers in silver chloride for temperatures down to −180°C so that the failure cannot be attributed to non-absorption of the red light.

Energy Relations in the Resonators

The break-up of colloidal silver particles into smaller and more numerous parts which is observed to occur on illumination of photo-chloride with red light is supposed to be due to the optical resonance of the particles to the incident light with consequent absorption of energy and ejection of electrons. This is a somewhat surprising combination of classical and quantum mechanisms and requires further consideration.

It was thought that the problem might be approached by considering the energy levels of the free electrons in the metallic lattice. Certainly these levels depend upon the dimensions of the particle but calculation of the approximate size of particle which might be expected to have the correct energy levels necessary to explain the effect appears to lead to dimensions of entirely the wrong order of magnitude—only 20 or 30A in length. The particles of silver are actually of the order of magnitude of a wave-length of light so that this approach to the problem is clearly inadequate.

The work function of a metal surface, or the work which has to be done in order to extract an electron, is due in part to the image force, i.e., the force of electrostatic attraction of a conductor for a charge. The force of attraction is $-e^2/4kx^2$, where x is the distance of the charge e from the conducting surface and k is the dielectric constant of the surrounding medium. For silver chloride k is 11.2 so that the image force on an electron escaping from silver into silver chloride is less than a tenth of that in vacuum. Another way of regarding this would be to consider the periodic field of the silver chloride lattice as producing a Schottky effect, or decrease of the work function as a result of the applied electric field.[6]

Thus, whatever the viewpoint, the energy which must be communicated to an electron in the silver before it can be ejected into the silver chloride lattice may be expected to be considerably less than the photoelectric threshold energy in vacuum, which for silver is about 3.8 electron volts and corresponds to a wave-length of 3390A. It is necessary to assume that the threshold wave-length is displaced in silver chloride into the red end of the visible spectrum.

The probability of ejection of a photoelectron by light of shorter wave-length than the threshold depends upon the intensity, i.e., upon the square of the amplitude of the electric vector at the surface of the metal. Now, at resonance the electric field at the surface of the resonator is many times the electric field in the incident wave. In Hertz's experiments the field at resonance exceeded the breakdown potential and sparks were seen to jump the spark gap. The probability of emission of electrons is therefore a maximum at the resonance frequency and those particles which are of suitable size and orientation to resonate to the incident light eject photoelectrons into the silver halide much more abundantly than the others.

In conclusion it may be said that photo-adaptation of a silver-silver chloride system is due to absorption of light by the particles of silver in the silver halide lattice. Only those particles which are of the right linear dimensions and of correct orientation can respond to the incident light. Whereas in colloidal suspensions of silver the effect of resonance is to scatter the light, in the silver halide lattice the silver particles can absorb the light and eject an electron, as a result of the peculiar boundary conditions. This ejection of an electron leaves behind a silver ion and reforms a silver atom in another part of the lattice. Migration of the chloride ion balances this redistribution of ionic silver and the result of the whole process is that those silver particles which absorb red light of the particular plane of polarization are ultimately converted into smaller particles which resonate to and absorb light of

[26] Hilsch and Pohl, Zeits. f. Physik **64**, 606 (1930).

shorter wave-length. Consequently, the silver image is color-adapted, dichroic and because of the relationship between refractive index and absorption is also doubly refracting.

These properties of the image persist so long as the character and orientation of the silver particles remains unchanged, even if the surrounding medium be changed. However, if the medium be altered the absorption curves are displaced along the wave-length scale in the ratio of the change of refractive index.

This picture of the effect is believed to be more satisfactory than those previously proposed which involve unwarranted assumptions as to micellar structure and deformation and which adequately explain neither the optics nor the physics of the process. It assumes nothing but the experimental observations that both silver and silver halide are necessary and sufficient for the production of the effects and that the effects once produced are properties only of the silver in the image.

Summary

(1) Red light causes a decrease in the size and an increase in the number of silver resonators present in a photo-halide emulsion.

(2) The photo-adapted image produced in a thick cell of photo-halide gel by unpolarized red light is dichroic when viewed perpendicular to the path of the beam.

(3) Low temperatures inhibit the formation of a photo-adapted image.

(4) A mechanism of photo-adaptation is postulated which assumes ejection of electrons from the silver resonators, these electrons reforming silver from silver ion at another point in the crystal lattice. The lattice is reformed by the migration of chloride ion. Only correctly dimensioned and oriented particles can respond to the incident polarized light.

Acknowledgment

Our grateful thanks are due to the National Research Council for a Fellowship to one of us (A. E. C.) which made this work possible and to Dr. S. E. Sheppard of the Eastman Kodak Company Research Laboratory for his sustained interest and encouragement.

Solarization at Low Intensity

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(Received June 27, 1934)

INTRODUCTION

THE peculiar phenomenon of photographic reversal has been the subject of much discussion. It has been found, as related in the abundant literature on the subject, that with progressive exposure the developability of the latent image, after increasing to a maximum, falls with prolonged exposure. The portion of the Hurter-Driffield characteristic curve which displays this phenomenon is called the region of solarization. In this portion of the curve, Trivelli[1] distinguishes: (a) a region of the first reversal of the solarization, (b) a region of minimum density, and (c) a region of the second reversal of the solarization, which is a re-reversal. What happens to the solarization curve if the exposure time is extended beyond that which is necessary to obtain the second reversal mentioned above? Observing Trivelli's solarization curve,[2] it may be noted, that in the region of the second reversal the density of the latent image is increasing, therefore the curve is bending upward. Now the question is will the density increase to a maximum value and retain that value or will the curve, beyond the second reversal, contain other maximum and minimum points similar to the ones established by Trivelli?

Several theories have been suggested to explain the first reversal of the solarization. Trivelli[3] divides these into two groups: (a) theories which try to explain the solarization as a development effect, and (b) theories which try to explain the solarization as a pure latent image effect. At present, we have two latent image theories of the solarization. These theories are proposed by two groups of investigators. The first group, which includes Lüppo-Cramer,[4] M. Andressen,[5] J. M. Eder,[6] C. E. K. Mees,[7] W. Abney,[8] H. S. Allen[9] and H. Kieser,[10] proposed the regression theory, i.e., they consider the action of halogen liberated from the silver halide of the grain on the centers of development as the cause of the first reversal of the solarization. In the second group we find such men as J. Eggert,[11] W. Noddack,[12] H. Arens,[13] H. Scheffers,[14] W. Leszynski[15] and W. Meidinger,[16] who defend the progression theory, i.e., they regard the cause of the first reversal of the solarization as a coagulation of the photochemically formed silver on the surface of the silver halide grain.

According to the concentration-speck theory,[17] which is included under the regression theory, the result of the photochemically liberated halogen is an increase in the size of the speck until a maximum size is reached. After this maximum value is obtained the size of the speck diminishes on account of the recombination of liberated bromine thus giving rise to the first reversal. When the concentration action of the speck is exhausted, it acts further only as a spot of disturbance in the crystal lattice, which grows slowly during continued exposure. At the same time the photochemically liberated halogen diffuses away through the gelatin. Thus we have, in the region of solarization, two reactions taking place in opposite directions. The first, which dominates, is tending to diminish the size of the speck through increasing pressure of the halogen, which is no longer absorbed by the silver sulphide. The second, which is not so pronounced, is endeavoring to increase the size of the speck by the printing-out decomposition. Therefore the size of the speck will go through a minimum, after which the second reversal of the solarization appears.

[1] Trivelli, J. Frank. Inst. **209**, 373 (1930).
[2] Trivelli, J. Frank. Inst. **209**, 46 (1930).
[3] Trivelli, J. Frank. Inst. **209**, 374 (1930).
[4] Lüppo-Cramer, Zeits. f. wiss. Phot. **25**, 224–308 (1928).
[5] M. Andressen, Das latente Lichtbild, 36 (1913).
[6] J. M. Eder, Zeits. f. physik. Chemie **117**, 293 (1925).
[7] C. E. K. Mees, Abridged Sci. Pub. **1**, 74 (1913–14).
[8] W. Abney, Phot. J. **48**, 318 (1908).
[9] H. S. Allen, *Photoelectricity*, 1913.
[10] H. Keiser, Zeits. f. wiss. Phot. **26**, 321 (1929).
[11] J. Eggert, Eder's Handb. **21**, 246 (1927).
[12] W. Noddack, Eder's Handb. **21**, 246 (1927).
[13] H. Arens, Zeits. f. physik. Chemie **114**, 337 (1925).
[14] H. Scheffers, Zeits. f. Physik **20**, 109 (1929).
[15] W. Leszynski, Zeits. f. wiss. Phot. **24**, 282 (1926); Phot. Ind. **26**, 425 (1928).
[16] W. Meidinger, Handb. d. Phys. Optik. 50 (1927).
[17] Trivelli, J. Frank. Inst. **209**, 387 (1930).

This second reversal[18] has been obtained in various ways; for example, with light, hydrogen-peroxide, ozone, persulfates, sodium arsenite, stannous chloride, with certain organic peroxides and other fogging agents. Here it is well to note that none of these experiments have shown reversals of an order higher than the second, although the probability of their existence has been admitted by Lüppo-Cramer.[19] The details of these experiments will not be discussed in this paper since none of these methods was employed in our experimentation.

H. Arens,[20] in his paper entitled *The Properties of Photographic Reversal Phenomena*, recognizes three types of photographic reversal. In the first type, the exposure to light causes the formation of finely-deposited silver, which is very reactive to the developer and forms the normal latent image. This silver acts catalytically on the development of additional silver salt. With further exposure the silver becomes compact by aggregation and loses its catalytic properties. The essential characteristic of the second type is the oxidation by light of the silver of the granules in the presence of Br molecules or ions. The third type is based on the diffusion of the bromine, which is liberated by the action of light, from the interior of the granules, and which, on reaching the surface, reacts again with the silver, thereby reversing the original photographic reduction.

From his extensive study of the solarized latent image, especially with regard to physical development, H. Arens[21] has been able to contribute greatly to the literature of this field. A few of the experimental facts stressed in some of his papers may be briefly expressed as follows:

(1) A large quantity of potassium bromide present during peptization produces a raising of the minimum of the solarization curve. Also K Br or Br-ions present during the illumination of the plate increases the solarization.[21]

(2) Silver-equivalent bromide bodies peptized with ammonia show strong solarization, which increases with increasing ammonia content.[22]

(3) In the case of peptized silver-bromide emulsions the phenomenon of solarization requires that in this region the size of the particles should continue to increase, but their number to diminish.[23]

(4) An emulsion which is prepared shortly after the separation of the peptized silver compound, is either not solarized at all or only very slightly affected. If the Ag-Br precipitate is allowed to stand for some time the property of solarization increases in an emulsion prepared from such material. When the silver ions are reduced as far as possible by the addition of K Br, a Ag Br is obtained which solarizes strongly after peptization.[24]

(5) A special emulsion obtained by preparing Ag Br in the absence of gelatin and then dispersing it in the latter shows that the total photolytic silver increases progressively with exposure throughout the solarization and second inversion regions, but the number of development centers declines after passing through a maximum. Thus solarization is to be ascribed to a regression in the number of centers rather than in the quantity of photolytic silver.[25]

(6) Solarization either appears for both primary and secondary physical development or it does not. With primary development it is more pronounced than with secondary. And generally, emulsions showing solarization with chemical development will show it with physical development, the effect being more marked for chemical development.[26]

The micelle theory of the latent image, as presented by F. Weigert,[27] holds that the micelles, which represent the latent image, are made up of silver halide, water, gelatin and silver, and they possess, when illuminated, an internal local energy accumulation. During the development of the latent image the molecules of the developer are activated by this supply of internal energy resulting in a chemical intra-micelle process. The *S*-shape of the blackening

[18] Trivelli, J. Frank. Inst. **209**, 374 (1930).
[19] Lüppo-Cramer, Das latente Bild, 37 (1914).
[20] H. Arens, Zeits. f. wiss. Phot. **11**, 337–386 (1925).
[21] H. Arens, Zeits. f. wiss. Phot. **28**, 91–110 (1930).
[22] H. Arens, Zeits f. wiss. Phot. **28**, 91–110 (1930).
[23] H. Arens, and K. Kipphan, Zeits. f. wiss. Phot. **28**, 229–238 (1930).
[24] H. Arens, Zeits. f. wiss. Phot. **28**, 341–48 (1931).
[25] H. Arens, Zeits. f. wiss. Phot. **30**, 127–40 (1931).
[26] H. Arens, Zeits. f. wiss. Phot. **31**, 317–23 (1933).
[27] F. Weigert, Zeits. f. wiss. Phot. **29**, 191–201 (1930).

curve may be accounted for by this reaction and the formation of metallic silver which causes a reduction of the available energy and consequently a decrease in the density of the latent image.

V. P. Lubovich,[28] a staunch believer in the second and higher order reversals concludes from his experimentation that for each kind of emulsion, at a given wave-length and intensity, there is a definite time of exposure for maximum density of the developed image. If the time of exposure exceeds this interval the image will decrease in density and may become entirely indistinguishable from the general background. One of his experimental curves shows the second reversal together with the beginning of the third. From this, he deduces his idea that the process of repeated reversion is analogous to a damped vibration.

A totally different theory is put forth by W. Leszynski.[29] According to him, if one exposes an emulsion behind a grey-wedge, without pre-exposure, the second reversal is absent. If, on the contrary, the emulsion is first pre-exposed diffusely to the point of solarization and then the exposure continued behind the grey-wedge, one obtains a renewed rise of the density up to a second maximum. A similar experiment was performed without the wedge, and the same results were obtained. Therefore Leszynski concluded that "if the exposure of the solarized plate was interrupted there appeared after the interruption the second reversal, which was missing when the interruption did not occur. It is borne out therefore that the interruption is the deciding factor for the second reversal."

It is the purpose of this paper to present the results obtained during a period of two and a half years of constant experimentation in the region of solarization. The order of the reversals attained is higher and the method for obtaining them is different from any heretofore mentioned in the literature on the solarized latent image; hence no parallel experiment can be cited. Therefore the remaining part of the paper will be confined to the presentation and discussion of the experimental data.

[28] V. P. Lubovich, J. Opt. Soc. Am. **14**, 309 (1927).
[29] W. Leszynski, Phot. Ind. **28**, 1412–16 (1930).

Apparatus

The camera used in this experimentation is shown diagrammatically in Fig. 1. It is 460 cm long, 126.5 cm wide and 126.5 cm high. The plate holder *P*, housed in a special compartment at the rear of the camera, is divided into four sections. Each section is capable of holding one 9×12 cm plate. Thus it was possible to expose four photographic plates at the same time, under the same conditions. Referring to Fig. 1, at the extreme right, it will be noted that there are 5 rows of 11 small black circles in each of the two sections shown. These circles represent holes in a metal plate which is fixed directly in front of and in contact with the plate holders. Light was permitted to pass through these openings, which have a diameter of 0.812 cm, and fall upon the photographic plate giving a circular image. A metal strip covered each row to prevent undesired exposure to light. The openings were uncovered in order of succession beginning with the inner edge of the top row of each plate and working outward with each movement of their respective covering strips. *A*, *B*, *C* and *D* are four diaphragms, with circular openings, placed at intervals inside the camera in order to prevent internal reflections. The entire inner surface of the camera together with both faces of the diaphragms were coated with lampblack. "O" is a diaphragm shutter placed in the front of the camera to regulate the exposure both as to time and light intensity. In all exposures the opening at "*O*," which had a diameter of 0.5 cm was covered by a piece of photographic plate-glass ground on both sides with emery powder.

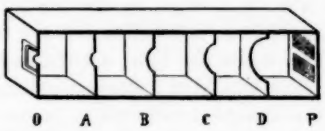

FIG. 1. Diagrammatical representation of the camera employed in this experimentation.

The sources of light were, in turn, a 400 watt 115 volt, and a 1000 watt 115 volt, well seasoned, Mazda lamp. These were burned at a constant voltage of 108 volts. They were placed 40 cm in front of the opening *O*, in such a way that the emulsion was not exposed to the direct light of

the filament. This was done in order to eliminate pinhole image effects.

The approximate intensities which fell upon the plates were found to be 0.00012 and 0.0003 footcandle. These values were determined by means of a General Electric Footcandle Meter and the inverse square law formula, $I : I_1 = D_1^2 : D^2$.

The current was obtained from a direct-current compound-wound generator, with manual field control. Accurate timing of exposures was ensured by placing a relay in the control circuit, in such a manner as to start and stop a telechron electric clock when the light was turned on and off. The relay was also arranged to ring a bell if the light should accidentally go off during the exposure. This gave a security against the possible uncertainty in the time of exposure which would have been introduced by the burning out of a fuse or a lamp, or the stoppage of the city current supply.

Before proceeding with the description of the experimentation the authors wish to emphasize the fact that all the curves shown in this paper were obtained from plates which were exposed continuously to the source of light. No interruption occurred in the exposure once it was begun. During the *preliminary* experiments, however, several interruptions were made in the exposure of the plates. These interruptions had no outstanding effect on the shape of the curve, since the same number of reversals were obtained whether or not the interruption occurred. This is in contradiction to the hypothesis of Leszynski, who attributes the existence of the second reversal to the interruption in the exposure.

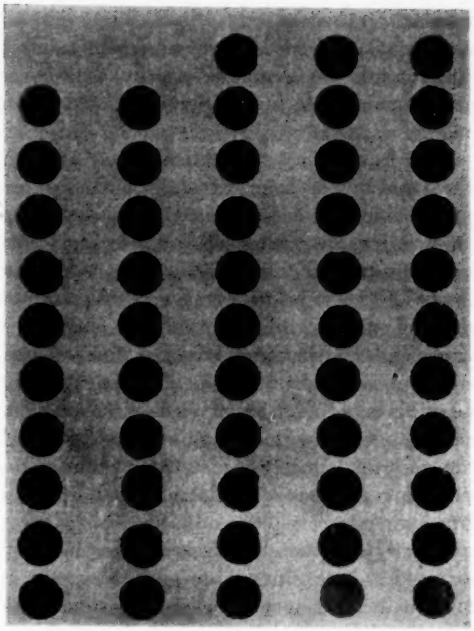

FIG. 2. The plate reproduced in the above figure shows the arrangement of the exposures on the photographic plate which was employed in this experimentation. Studying the figure one observes that all the exposures, except the first one in the upper left-hand corner of the second row, are apparently of the same density. This is true since it is a reproduction of one of the four plates whose average curve is displayed in Fig. 6 and not a reproduction of one of the reversal plates.

MATERIAL AND DEVELOPMENT

The material employed in this experiment was limited to Extra Fast, Dry, Undyed Hammer Plates.[30] By reason of the long period of time over which this experimental work was extended, it was impossible to have all the plates of the same emulsion number. Thus during the course of the work plates of various ages were employed, the oldest being 621 days old. The results obtained from these several plates agreed as to the number of reversals obtained during a given time of exposure. This is a point in favor of the accuracy of the results displayed in this paper. Similar data collected from plates all of the same type but differing in emulsion number are more convincing than data obtained from plates having the same emulsion number. Table I has been prepared to show the age, at the beginning

[30] We wish to thank the Hammer Plate Company of St. Louis, Missouri for the plates and their hearty cooperation.

TABLE I.

Emulsion number	Date of coating	Dates of exposure	Age	Figure number
4633	March 17, 1931	Nov. 28–Dec. 19, 1932	621 days	3 Curve *c*
5063	July 12, 1932	Nov. 28–Dec. 19, 1932	139 days	3 Curve *a*
5063	July 12, 1932	Nov. 28–Dec. 19, 1932	139 days	3 Curve *b*
5063	July 12, 1932	Nov. 28–Dec. 19, 1932	139 days	4 Curve *d*
5063	July 12, 1932	Jan. 9–Mar. 23, 1933	181 days	4 Curve *e*
5252	Feb. 9, 1933	Mar. 27–Apr. 15, 1933	46 days	5 Curve *f*
5252	Feb. 9, 1933	May 7, 1933	87 days	6
5400	July 26, 1933	Oct. 13–Nov. 1, 1933	79 days	5 Curve *g*
5400	July 26, 1933	Nov. 6–Nov. 25, 1933	103 days	5 Curve *h*
5400	July 26, 1933	Feb. 21–Mar. 12, 1934	210 days	5 Curve *i*

of the exposure, of all the plates whose data are shown graphically in this paper (Fig. 2).

All the plates were developed, for a period of five minutes, in the following developer which was kept at a temperature of 65±1°F.

Development formula		*Acid hypo fixing bath formula*	
Water	3632 cc	Water	240 cc
Hydrochinon	75 g	Sodium hyposulphite	60 g
Sodium sulphite	240 g		
Sulphuric acid	3.5 cc	Water	75 cc
Potassium carbonate	180 g	Sodium sulphite	15 g
Potassium bromide	15 g	Acetic acid (28%)	40 cc
		Powdered alum	15 g

During development the surface of the plates was brushed lightly with a camel's hair brush[31] in order to ensure uniform development. Each plate was washed, for one minute, in water and fixed in the above acid hypo solution. After an interval of 15 minutes the plate was removed and placed in running water where it remained for two hours.

The diffused densities were measured by means of the "differential photoelectric photometer" described by F. K. Richtmyer.[32] According to Richtmyer, differences in densities of 0.01 percent are measurable with this instrument. The accuracy of the results was checked several times, during the course of the work by remeasuring not only individual exposures but also entire plates with a different setting of the stationary lamp. In all cases the results obtained were in very close agreement.

The curves shown in Fig. 3 were obtained by exposing, simultaneously, three photographic plates to the ground-glass-source of light which, in this case, was illuminated by a 400 watt lamp. The longest exposure, as indicated by the log of exposure line 4.442, was 27,652 minutes. The plates from which curves *a* and *b* were obtained had the same emulsion number (5063) and when this exposure was begun 139 days had passed since they were coated. Curve *c* was procured from a plate whose emulsion was about four and a half times as old, 621 days having elapsed between the date of coating and the date of exposure.

Comparing curves *a*, *b* and *c* one observes, that the number of reversals in each case is the same; and that the various maximum and minimum points lie on approximately the same log of exposure lines. Curves *a* and *b* are very much alike especially in density values and the shape of the curves, which is to be expected since they were obtained from plates having the same emulsion number. Curve *c* lies wholly below *a* and *b* since its density is lower; furthermore the depths of its reversals[33] are less than those of *a* and *b*. Hence we would draw the conclusion, which has been confirmed by other curves not shown in this paper, that with the aging of the emulsion the solarization effect decreases. In passing, we wish to remark that the first four reversals displayed in the above figures, were perfectly reproduced with plates of various emulsion numbers including 5063 and 4633. These identical reproductions were also had in the case of time schedule rearrangements, all of which had the same total maximum time of exposure although a greater number of exposures were made during this time period.

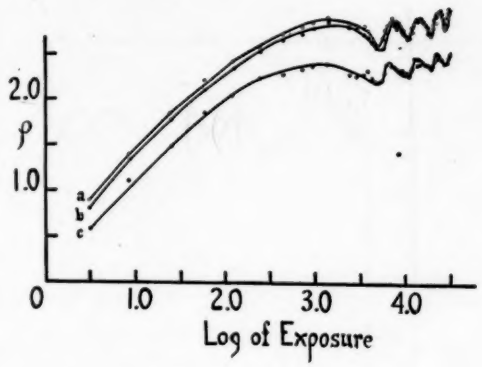

FIG. 3. Three curves showing eight reversals which were obtained from plates exposed simultaneously to the intensity provided by the 400 watt lamp for a period of 27,652 minutes. The plates from which curves *a* and *b* were obtained had the same emulsion number (5063). The plate employed in obtaining curve *c* was much older, its emulsion number being 4633.

Curve *d*, displayed in Fig. 4, is the result of averaging the respective densities of each exposure shown in curves *a* and *b* of Fig. 3 and then plotting this average against exposure-time in hours. Here the relation between the actual time required to produce the reversals is more

[31] W. Clark, Phot. J. 76, Feb. (1925).

[32] F. K. Richtmyer, Phys. Rev. 6, 66 (1915).

[33] By the term " depth of reversal " we mean the interval between a maximum and the following minimum.

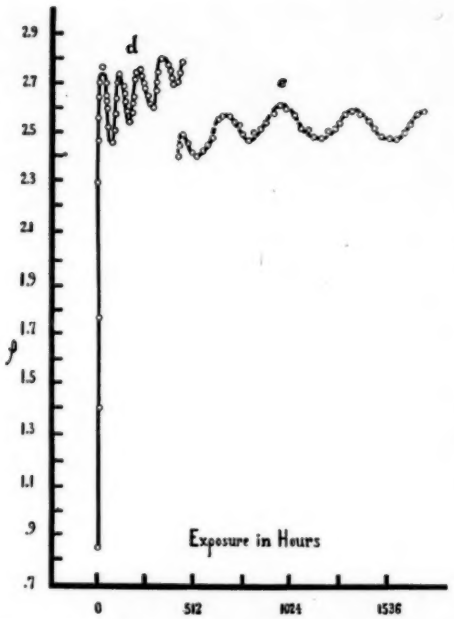

FIG. 4. Curve *d* represents the average of curves *a* and *b* in Fig. 3 plotted against exposure in hours. Curve *e*, which was obtained from plates having emulsion number 5063, is the result of further exposure to the intensity provided by the 400 watt lamp.

obvious and one readily sees that the time between two successive maxima is progressively longer as the order of the reversals increases.

Curve *e*, exhibited here, is an average curve made from a set of four plates which were exposed simultaneously. Although the emulsion number of the plates used was the same as for curves *a* and *b* the average curve obtained was displaced in the negative direction with respect to the density axis by an amount equal to 0.3. If this amount is added to the relative densities, which are represented as points on curve *e*, one observes that the first three points on *e* will coincide with the last three points on *d*. Theoretically, this coincidence is to be expected, since the same times of exposure and same emulsion number were employed in each case. If these two curves were actually joined together the completed curve $(e+d)$ would show 16 reversals which were obtained during a period of exposure equal to 1758 hours and 52 minutes. Thus the investigation of the region of solarization has been extended beyond the log of exposure line 4.442. The dotted portion is similar to the full lined in that the time between two successive maxima or minima is increasing and the depth of the reversals is decreasing. This fact bears out the damped vibration analogy of Lubovich which is mentioned in the introduction; it also indicates that the centers of development are produced with greater difficulty.

The four average curves,[34] exhibited in Fig. 5, were obtained by exposing, in each case, four photographic plates to the ground-glass-source of light which was illuminated by a 1000 watt Mazda lamp. Actual measurement showed that the light incident upon the plates was two and a half times as great as that which resulted from the 400 watt lamp. The time schedule used for the plates from which curve *f* was obtained was slightly different from the one used for curves *g*, *h* and *i*. Curve *f* having 300 minutes of exposure less than curves *g*, *h* and *i*. This amount was small when compared with the total time of 26,753 minutes, and as is to be expected, no appreciable effect can be noted. Also the emulsion number (5252) of the plates used for curve *f* differed from the emulsion number (5400) of the plates used for curves *g*, *h* and *i*. The ages of the emulsions when exposure was begun varied

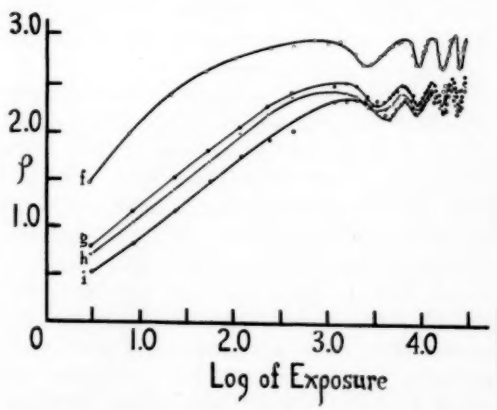

FIG. 5. Reversal curves obtained from exposure to light intensity provided by the 1000 watt lamp. The plates from which curve *f* was obtained had an emulsion number 5252, while the plates from which the other three curves were obtained had the emulsion number 5400.

[34] These curves were obtained by averaging the respective densities of the corresponding exposures on a set of four plates which were exposed simultaneously.

as follows: curve *f*—46 days; curve *g*—79 days; curve *h*—103 days and curve *i*—210 days. In this case the difference in age is not as great as it was in the curves displayed in Fig. 3, consequently the effect of age will not be so well marked. However on close examination, one observes that these four curves have a decrease both in density and in the depth of the reversals. Here it is well to note that this difference may be due to other causes, for instance, a flaw in the emulsion, or a decrease in intensity resulting from the aging of the filament of the Mazda lamp. But after considering various experimental facts the authors are inclined to believe that the aging of the emulsion is chiefly responsible for these changes. Although the curve *h* has the same expcsure time and emulsion number as *g* and *i*, it differs from them in another respect. Before beginning the exposure which gave this curve, the plate holder at the back of the camera was rotated through 90°. Thus the four plates were in a new position with respect to the incident light beam. The curve obtained from these four plates does not differ greatly from curve *i*, which was the result of an exposure made immediately after the one for curve *h* with the plate holder returned to its original position. Thus one observes that the experimental arrangements for curves *h* and *i* differ chiefly in the position of the plate holder and slightly in the age of the emulsion.

Sources of Error

The following sources of possible error have been examined:

(1) variations in source of light due to change in voltage
(2) non-uniform distribution of intensity within the camera
(3) uneven coating of the photographic plate
(4) variation in exposure time
(5) variation in temperature of the developer
(6) non-uniform development
(7) error in photometric measurements.

Each of these sources of error was examined by means of a series of experimental checks.

The voltage applied to the lamp was checked several times during the day exposures and the variation, if any, was less than ± 1 volt. During the night exposures the voltage was rarely checked, especially during the hours between midnight and 6 A.M. Thus if there was a systematic variation of the light intensity at night, it seems probable that the maximum and minimum points on the curve would be effected. To test this the amount of day and of night exposure for each point on the curve was summed up. No correlation was found to exist between the excess day and night exposure and the position of the maximum and minimum points. Therefore it was concluded that the voltage was as constant during the night exposures as it was during the day exposures.

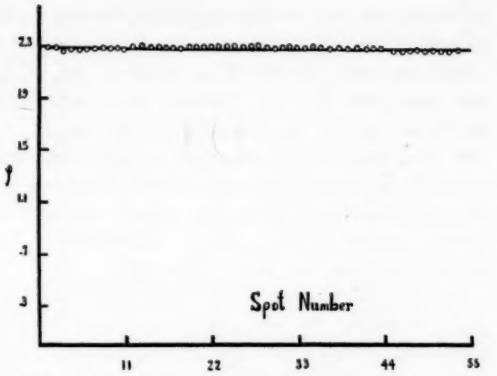

Fig. 6. This curve displays the distribution of intensity inside the camera over the region occupied by the plate holders. It was obtained from plates which had an emulsion number 5400.

One of the most important features of the entire experiment is the distribution of the light intensity within the camera. This was investigated by placing four plates in the plate holder, having all exposure holes open, and exposing them simultaneously for ten minutes, to the intensity supplied by the 1000 watt lamp. The average curve obtained from these plates is displayed in Fig. 6. The deviation of the points from a straight line is slight, therefore it was concluded that the maximum and minimum points of the solarization curve were not the result of shadows, reflections or pinhole images. This conclusion was confirmed by a second experiment. Before proceeding with this experiment the plate holder was rotated through 90°, then the four plates were exposed, progressively, to the intensity supplied by the 1000 watt lamp. The average curve obtained from these four plates, which were exposed in a position at right angles to the original position, is displayed

as curve h in Fig. 5. The similarity of this curve to others, which were made under the original conditions, has already been pointed out. Next the position of the various maximum and minimum points on the plate with respect to row and number of exposure in the row was studied. No correlation was found to exist since the maximum and minimum points occurred on various parts of the plate, consequently no definite pattern was being followed.

An uneven coating of the photographic plate could possibly be responsible for the results displayed in this paper but after considering the number of curves obtained during this experimentation and especially the curve exhibited in Fig. 6, it seemed to be a very unlikely source of error. The slight deviation of the points from a straight line, which is noted in this figure, portrays the regularity of the emulsion coating.

As previously noted in this paper, the accurate timing of exposures was ensured by the relay-clock-bell system which was placed in the control circuit.

During the development of the photographic plates special attention was given to the temperature of the developing solution. The plates were placed in the developer when the thermometer registered 65°F. At the end of the development period the temperature was again noted and if there was a change it never exceeded +1°F. During warm weather the developer tray was surrounded with ice to guard against a rise in temperature.

Uniform development, as mentioned above, was obtained by brushing the surface of the plate, while it was in the developer, with a camel's hair brush.

Finally the experimental error in the photometric measurements was calculated by taking nine readings of one exposure, the readings being made at intervals of seven minutes. This particular time was chosen since approximately one hour was required to measure one of the experimental plates. The probable error was calculated by means of the following formula:

$$PE_s = 0.6745 s/n^{\frac{1}{2}}$$

where s = the mean deviation of the readings, and n = the number of readings made. This gave a probable error in density measurements of 0.00088.

The result of our experimentation may be briefly summed up in two conclusions:

(1) The solarization curve, beyond the second reversal, contains a set of maximum and minimum points.

(2) The aging of the emulsion decreases both the density resulting from the development of the latent image and the solarization effect.

Finally it must be noted that there is considerable room for further study in this field before comprehensive conclusions can be drawn. For example the experiment described in this paper could be repeated using different types of photographic material. J. M. Eder[35] states that the threshold value of the first reversal varies with the photographic material.

The authors wish to express their appreciation to Dr. F. E. Poindexter, who suggested this experimentation; to Rev. J. I. Shannon, S. J. for his interest and valuable suggestions; and to Mr. V. P. Jacobsmeyer, S. J. and Mr. J. V. Downey, S. J. for their assistance in changing the night exposures during the past year.

[35] J. M. Eder, Phot. 645 (1902).

Transparency of Thin Metallic Films in the Ultraviolet

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(Received August 15, 1934)

A technique for investigating the transparency of metallic films from the visible down to 1300A is described and results are given for ten different metals investigated. No new strong regions of transparency were discovered.

THIS work was undertaken with the idea of searching for possible transparent regions in thin films of as large a number of metals as possible down to 1300A. The measurements were made photographically and the results are given in terms of the percent of light transmitted by a film of given thickness. This information is not sufficient for the determination of absorption coefficients, since the reflecting power also varies greatly, and also because the Schwarzschild coefficient is comparatively unknown at these wave-lengths, but for a first survey it was felt that general information as to any possible "transparent" regions would be of value. The following metals have been investigated: Cu, Ag, Au, Mg, Al, Sn, Pb, As, Sb, and Bi; and while all transmit some light at the thicknesses used, in no case has any very marked transparency been discovered.

The vacuum prism spectrograph employed is that described by Cario and Schmidt-Ott.[1] The optical parts are of fluorite of quite small dimensions; Cramer contrast plates sensitized with Nujol were used. The absorbing films were placed about halfway between the slit and the collimating lens. The source of light was a water-cooled hydrogen discharge tube run by means of a 2200 volt transformer taking 6 amperes in the primary from the 220 volt mains. The tube was made of Pyrex glass with aluminum electrodes in the form of hollow cylinders 4.5 cm in diameter and 10 cm in length mounted in side tubes. The length of the discharge tube was 30 cm and its inside diameter 0.8 cm; it was viewed end-on and its inner surface was lightly silvered to prevent the appearance of the atomic spectrum of hydrogen.

For obtaining metal films of the necessary thinness, various methods that have been used before, such as those of Rupp[2] and G. P. Thomson,[3] were tried without conspicuous success, and the following procedure was then found easier and apparently quite satisfactory.

The metals were deposited by evaporation on a thin film of celluloid which was prepared by the method described by O'Bryan.[4] A two percent solution of celluloid in amyl acetate was made and one drop of the solution was allowed to fall on a clean water surface. The solution spread rapidly, forming a film from 10 to 12 cm in diameter. The thickness of the films used varied from 200 to 400A. After the amyl acetate had completely evaporated a small portion of the film was put on a flat brass ring 2.2 cm outside diameter and 0.5 cm inside diameter. The film was attached by lowering the brass ring, held by tweezers in a horizontal position, until it just touched the surface. Then the film around the ring was cut away and the ring with the adhering film was raised from the surface of the water. The excess water was carefully taken off by means of filter paper and the film allowed to dry. The process was repeated once; the two films of celluloid adhered together very well and made a much more satisfactory base than one single thicker film.

After some attempts with sputtering, the method of evaporation in a vacuum was adopted, and proved satisfactory for a number of metals. The arrangement as described[5] by Cartwright and Strong, with certain modifications, was used. The metal to be deposited was placed in a small

[1] G. Cario and Schmidt-Ott, Zeits. f. Physik **69**, 719 (1931).

[2] E. Rupp, Ann. d. Physik **85**, 981 (1928).

[3] G. P. Thomson, Proc. Roy. Soc. **A125**, 352 (1929).

[4] H. M. O'Bryan, J. O. S. A. **22**, 739 (1932).

[5] C. H. Cartwright and John Strong, Rev. Sci. Inst. **2**, 189 (1931).

vertical helix of tungsten wire 0.013 cm in diameter. A small stand having devices for holding three celluloid-coated brass rings was placed directly over the tungsten coil. The whole was placed under a bell jar and evacuated. A charcoal tube was placed immediately below the opening in the brass plate and from there connection through an additional simple liquid air trap was made to the pumps. The evaporation of the various metals was effected by passing from one to four amperes through the coil. Before evaporation an electric heater was placed around the charcoal tube and the tube was baked for some time with the pumps running. The evaporation was started after the charcoal was immersed in liquid air and a "black" vacuum had been obtained. The amount of deposit was judged by viewing the films through the bell jar. By the above-described procedure the following metals were successfully evaporated: Cu, Ag, Au, Mg, Sn, Pb, As, Sb and Bi. In order to evaporate aluminum a slight modification[6] had to be made since at high temperatures aluminum reacts with tungsten and the coil burns out before a deposit is obtained.

The thickness of the metal films was calculated from the density and weight of metal deposited, which was obtained by weighing the ring and celluloid, before and after evaporation of the metal, on a microbalance. The possibility that the weight of the celluloid backing changed during evaporation was tested by weighing a number of celluloid films, then placing them over the tungsten coil in the bell jar, evacuating the bell jar and heating the coil with no metal in it. They were then taken out and reweighed and in no instance did they show an appreciable change in weight. No films were used that did not appear perfectly uniform by transmitted and reflected light.

The balance used had a maximum carrying capacity of twenty grams and a sensitivity of one microgram per tenth of a scale division. The weights of the films used varied from 20 to 400 micrograms.

The spectrograph was calibrated with a quartz mercury lamp, an aluminum spark in air, a vacuum carbon arc lamp, and a vacuum cadmium arc. A preliminary test of the transparency of the celluloid films showed that their absorption, though fairly small, was too great to be neglected, especially in the extreme ultraviolet. The apparatus was therefore arranged so that exposures could be made either through a metal on celluloid film or through a plain celluloid film, and several comparison spectra of this sort were placed on each plate.

Before taking exposures the spectrograph was evacuated until a green fluorescent vacuum was obtained. Hydrogen was then allowed to enter through a palladium tube until the pressure was such that a dark space of about 1.5 cm was present in the discharge tube. Exposures were made with a magnetically controlled shutter; the times ranged from 3 seconds to 45 minutes, and the discharge tube was run for about 10 minutes before the first one on each plate, to produce steady conditions. After the exposures were made the plates were bathed in acetone to remove the Nujol and then developed. By means of a Hartmann microphotometer the densities at 1608, 2000, 3100 and 4300A for each of the seven exposures on each plate were measured.

Results

The results are given in Table I, the numbers recorded being the percent of light transmitted by a film of given thickness at a specified wavelength. The number in parentheses in each case is an indication of the relative weight of the observation; it is simply the number of independent calculations from which the transparency was obtained. Individual determinations were not very concordant, and where this number is small the figure given must be regarded as purely qualitative. It is clear, however, that for none of the metals investigated is there any band resembling the well-known silver one. The most cursory inspection of the plates shows that this is not due to the choice of the particular wavelengths used for measurements. The experimental set-up was not arranged to measure the percent of light reflected in each case, and since this has been shown to vary not merely with wave-length, but even with the thickness of the

[6] John Strong, Phys. Rev. **43**, 498 (1933).

TABLE I.

Thickness of film (A)	Percent of light transmitted (and No. of measurements) 4300A	3100A	2000A	1608A
		Copper		
190	59 (2)	25 (8)	7.7(4)	4.2(4)
320	12 (3)	7.1(12)	4.3(5)	3.4(3)
740	8.6(6)	3.7(12)	—	—
940	4.2(6)	2.3(12)	—	—
		Silver		
170	43 (2)	37 (3)	21 (2)	24 (4)
250	11 (3)	32 (4)	7.7(2)	11 (3)
280	9.1(5)	42 (13)	7.1(1)	11 (1)
430	4.5(6)	32 (10)	4.3(2)	4.5(2)
590	0.5(1)	15 (6)	—	—
700	—	7.7(6)	—	—
		Gold		
60	50 (4)	45 (7)	50 (7)	37 (6)
95	37 (4)	36 (6)	30 (2)	23 (2)
110	36 (4)	29 (4)	22 (2)	14 (2)
140	26 (4)	22 (4)	20 (7)	11 (7)
210	22 (3)	15 (3)	15 (6)	5.1(6)
560	4.2(4)	2.0(16)	1.4(5)	0.5(2)
750	2.4(12)	1.1(16)	0.8(2)	—
		Magnesium		
310	18 (3)	25 (11)	21 (7)	9.5(4)
380	5.6(2)	7.3(6)	8.8(1)	—
580	2.4(3)	2.0(4)	2.9(2)	—
		Aluminum		
190	31 (5)	37 (16)	42 (5)	35 (5)
210	26 (2)	21 (3)	24 (2)	15 (2)
280	15 (2)	14 (3)	19 (2)	12 (2)
370	18 (2)	11 (3)	8.2(2)	2.6(1)
460	1.3(10)	1.1(10)	1.2(1)	—
940	0.3(3)	0.2(6)	—	—

Thickness of film (A)	Percent of light transmitted (and No. of measurements) 4300A	3100A	2000A	1608A
		Tin		
100	38 (2)	35 (3)	50 (3)	37 (3)
130	30 (3)	28 (8)	30 (2)	29 (2)
200	29 (2)	22 (2)	21 (2)	24 (2)
420	5.9(2)	3.7(5)	5.3(2)	5.3(2)
600	1.7(2)	1.3(2)	—	—
630	1.2(3)	0.7(6)	—	—
770	0.3(4)	0.2(4)	—	—
980	0.1(2)	0.1(2)	—	—
		Lead		
50	62 (1)	62 (6)	50 (3)	34 (3)
170	24 (2)	20 (4)	12 (2)	7.0(2)
250	24 (4)	11 (4)	5.3(2)	—
380	4.2(3)	2.8(7)	2.7(1)	—
550	1.7(6)	1.3(6)	—	—
		Arsenic		
100	26 (2)	24 (6)	31 (2)	28 (2)
150	23 (6)	21 (6)	17 (6)	16 (6)
270	7.8(6)	2.4(12)	0.9(2)	—
370	2.5(6)	0.5(6)	—	—
450	0.9(4)	0.3(5)	—	—
		Antimony		
100	24 (4)	25 (4)	18 (8)	14 (8)
130	25 (1)	22 (2)	16 (4)	14 (2)
230	6.5(2)	6.1(2)	7.1(4)	4.7(4)
270	6.0(2)	5.3(2)	4.2(4)	3.0(4)
360	0.7(4)	1.0(4)	0.8(2)	—
		Bismuth		
70	53 (1)	53 (2)	59 (2)	50 (3)
90	43 (1)	48 (4)	30 (2)	29 (2)
160	29 (1)	32 (2)	29 (2)	24 (2)
240	14 (2)	10 (4)	6.8(8)	5.4(6)
280	7.2(4)	8.3(4)	6.0(7)	4.3(8)
350	4.6(6)	5.4(6)	2.7(1)	—

Gold. The films appeared bluish green by transmitted light and characteristically yellow by reflected light.

Copper. The films appeared green by transmitted light and characteristically copper colored by reflected light.

Silver. Was quite easy to evaporate and always formed uniform films which appeared deep blue by transmitted light and characteristically silver colored by reflected light. It will be noted that very thin films are not especially transparent at 3100A, so that reflection is probably the main factor here, and is probably comparatively constant.

Magnesium. The films seemed to be very brittle. They appeared deep blue by transmitted light and gray by reflected light.

Aluminum. Uniform films were quite easily obtained when the special technique for evaporation was used. The films appeared blue by transmitted light and gray by reflected light.

Tin. The films appeared violet by transmitted light and gray by reflected light.

Lead. Appeared purple by transmitted light and gray by reflected light.

Arsenic. The films appeared orange by transmitted light and gray by reflected light. Earlier work on this metal with panchromatic plates showed that it had a maximum transmission band at 5700–6500A.

Antimony. Of all the substances tried antimony formed the best mirror surfaces. They appeared purple by transmitted light and gray by reflected light.

Bismuth. The films appeared blue by transmitted light and gray by reflected light.

film, at longer wave-lengths,[7] no calculations of absorption coefficients can be made.

This method of attack did not seem to be applicable to any other metals. Zn and Cd were evaporated repeatedly but the resulting films were always full of holes and under the microscope they exhibited a fern-like structure. An attempt was made to evaporate the following metals: Ca, Sr, Ba, Cr, Mo, W, Fe, Co, Ni, Mn and Pt, but in no case was a satisfactory deposit obtained.

The writer wishes to thank Dr. Robert d'E. Atkinson, who supervised the work of this experiment and through whose untiring efforts its completion was made possible.

[7] H. Murmann, Zeits. f. Physik **80**, 161 (1933).

An Indirect Method for Measuring Stray Light within the Human Eye

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(Received September 27, 1934)

THE source of stray light within the eye is three-fold, diffusion through the iris and the sclera, reflection from the retinal image, and scattering by the media. Diffusion through the iris and the sclera can be prevented by using an artificial pupil, but since stray light from the other two sources cannot be so easily avoided it is essential to develop a method for measuring it, so that one may be able to predict to what extent it comes into play in visual phenomena and in the electrical phenomena produced by applying a flash of light to the eye.

The authors have shown[3] that the changes in electrical potential produced by stimulating a rabbit's eye with light represents primarily the response of that part of the retina receiving only stray light. Techniques have already been developed for recording retinal action potentials from the human eye, and when this sort of thing comes to be used in the diagnosis of pathological conditions of the retina, a knowledge of the rôle played by stray light will be indispensable.

That stray light does play a measurable rôle in vision is suggested by Helson and Guilford's study of pigmentation of the eye.[4] They found that people with poorly pigmented eyes have a higher absolute threshold than people with strongly pigmented eyes, and they pointed out that this difference was attributable either to the difference in the quantity of stray light in the eyes or to the possibility that the pigment epithelium plays a direct rôle in vision.

Cobb[5] has also pointed out that scattered light must be taken into account in the understanding of visual phenomena.

Our method for measuring the stray light in the eye may be briefly stated as follows: A bright object was placed in the peripheral part of the visual field and the amount of stray light falling upon the fovea was measured by the change produced in the differential threshold. The change in threshold produced in this way can be compared with the change produced by illuminating the region of the fovea with a known intensity of light.

The apparatus we have used in our main experiments is illustrated in Fig. 1. The bright object in the periphery was provided by the illuminated aperture (3/8 in. in diameter) in the screen in front of lens I. Light from this object entered the eye after reflection by the right-angled prism. The image of the aperture was 14.5 in. away from the eye. The light from aperture I (1 mm in diameter) passed through lens I and came to a focus at the pupil of the eye. The right-angle prism, lens I and aperture I and the lamphouse were all mounted on a rigid frame which could be roughly adjusted to make the image of aperture I fall within the pupil. The position of aperture I could then be adjusted by

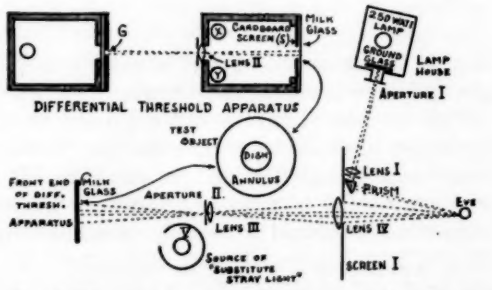

FIG. 1. Diagram of the apparatus for measuring the stray light within the eye. A bright object in the periphery is provided by the illuminated aperture in front of lens I whose image is reflected by the right-angle prism to the eye. A disk-annulus test-object for measuring the effect of stray light upon the threshold at the fovea is provided by the differential threshold apparatus. The observer does not see the test-object directly but an inverted image on the face of lens IV. The light falling on the front end of the differential threshold apparatus from source V serves as a substitute for stray light, and provides a standard with which the effects of stray light can be compared.

[1] The contribution of the senior author was made possible by a grant-in-aid from the Rockefeller Foundation for Research in Neurophysiology.

[2] National Research Fellow in the Biological Sciences.

[3] Work not yet published.

[4] H. Helson and J. P. Guilford, *The Relation of Visual Sensitivity to the Amount of Retinal Pigmentation*, J. Gen. Psych. **9**, 58–76 (1933).

[5] P. W. Cobb, *The Effect on Foveal Vision of Bright Surroundings*—II, Psych. Rev. **21**, 23–32 (1914).

screws in the vertical and horizontal dimensions so that its image could be made to fall exactly at the center of the pupil. This was done as follows: When the image was moved across the pupil the subject could tell when it reached the edge by the disappearance of the peripheral object. When it was known where to set the aperture to make the image fall on the opposite edges of the pupil, it could be calculated where to set the aperture to make the image fall at the center.

The visual angle which the peripheral object made with the fovea, could be varied but its distance from the eye was kept constant by a bar which hinged at one end with the frame holding the prism at a point directly below the prism, and which pivoted at the other end on a point directly below the eye. The head of the observer was fixed to a biting board.

The test-object employed in measuring the differential threshold at the fovea consisted of two parts, a central disk and a surrounding annulus. This test-object was provided by the differential threshold apparatus. The details of its construction and operation are as follows: The hole in the cardboard screen *S* allowed a disk-shaped area (9/16 in. in diameter) of the plate of opal-flashed glass to receive illumination from light sources *X* and *Y*. This illumination was kept constant. A circular spot (3/16 in. in diameter) at the center of this area received an additional illumination from *G*, since lens II focused an image of *G* on the milk glass. The observer could vary the intensity of this additional illumination by adjusting the size of the aperture behind lens II, by means of a flexible shaft. The observer's task consisted in increasing the brightness of the central disk from a subthreshold level until it became distinguishable from the annulus. The observer did not see the test object directly but an inverted image of the same size projected upon the face of lens IV, which was 16 in. from the eye. A spot at the center of the test object served as the fixation mark.

Aperture II (1 mm in diameter) was adjustable like aperture I so that its image could be made to fall exactly at the center of the pupil.

In order to provide some standard with which to compare the effect produced by the bright peripheral object upon the differential threshold at the fovea, a substitute for the stray light falling at the fovea was employed. This "substitute stray light" was provided by the light source *V*, which illuminated the front surface of the milk glass. This "substitute stray light" completely filled the aperture (2 in. in diameter) in screen I. The test-object which admixed itself to the center of this field, had a diameter of only 9/16 in. as compared with 2 in. for the total field.

We have specified the intensities of the peripheral object, the two parts of the test object, and the "substitute stray light" in terms of c per sq. ft. Since the stimulating capacity of an external object is a function not only of its brightness but also of the size of the beam entering the eye, it is perhaps necessary to point out that the beams of light entering the eye were all the same size, namely, 1 mm in diameter. Hence the brightness values we have used are all comparable with respect to stimulating capacity.

Fry served as subject in all the experiments. The right eye was used and the left eye shielded by a black card immediately in front of the eye.

One difficulty experienced with our apparatus was that part of the light which fell upon the eye from the peripheral object was reflected by the cornea, and was apt to pass through lens IV and come to a focus at the edge of aperture II, and reflect back to the eye, noticeably illuminating the face of lens IV. This difficulty was experienced when the visual angle between the fovea and the peripheral object was large, but it could be avoided by shifting the image of aperture I slightly to one side of the center of the pupil.

In our first experiment the effect of varying the brightness of the peripheral object upon the differential threshold at the fovea was investigated. The results are presented graphically in Fig. 2. The angle which the peripheral object subtended with the fovea was kept constant at 5°. The brightness of the annulus of the test object was kept constant at 0.05 c per sq. ft. The brightness values specified as differential thresholds represent differences between the brightness of the annulus and that of the central disk.

The measurements were all made at one sitting. The different levels of intensity of the peripheral object were taken in the order of increasing intensity. Alternate readings were made

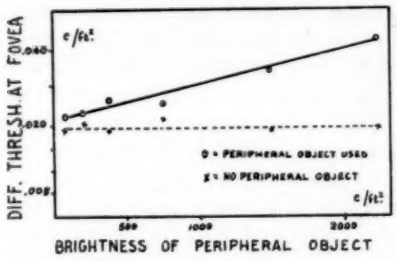

FIG. 2. The effect of increasing the stray light at the fovea upon the differential threshold.

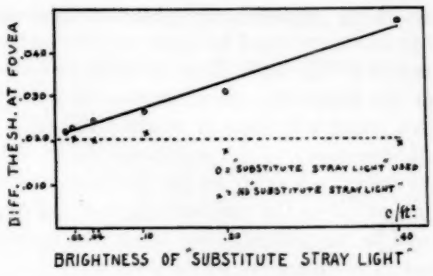

FIG. 3. The effect of increasing the "substitute stray light" upon the differential threshold.

with the peripheral object present and absent. Five such pairs of measurements were made at each level. The values given in the graph represent therefore averages of five readings. The circles represent the differential thresholds with the peripheral object present, and the crosses with the peripheral object absent. When the readings were alternated with the peripheral object absent and present as we have done, changes in the conditions of the eye can be detected, if any occur during the course of the experiment, by the change in threshold when the peripheral object is absent. This same general procedure has been used in all of our experiments.

In our next experiment the peripheral object was dispensed with and the effect of varying the "substitute stray light" upon the threshold was investigated. The results are presented in Fig. 3.

The brightness of the annular part of the test object was kept constant at 0.05 c per sq. ft. The different levels of intensity of the "substitute stray light" were taken in the order of increasing intensity, and the general procedure was the same as in the last experiment.

A comparison of the results will show that the intensity of the scattered light falling on fovea, when an object having a brightness of 2220 c per sq. ft. and subtending a visual angle of 1° 14′ projects its image upon the retina 5° from the fovea, is equivalent to an illumination coming from an external object whose brightness is 0.34 c per sq. ft. Since the beam of light entering the pupil is 1 mm in diameter, one can say that the intensity of the stray light is 2.87 photons.

The intensity of stray light which a bright object is capable of producing must be specified in terms of its brightness and the solid visual angle which it subtends and also in terms of the distance away from the retinal image at which the stray light is measured. In the above case, for instance, the peripheral light produces 0.00408 photons of stray light per c per sq. ft. of its brightness, and per unit of its solid visual angle (millisteradian) at a point 5° from the retinal image.

Our third experiment shows the effect of varying the displacement of the peripheral object from the fovea. The results are shown in Fig. 4. The brightness of the annular part of the test object was kept constant at 0.05 c per sq. ft. and the brightness of the peripheral object at 2220 c per sq. ft. The threshold-measurements for the different displacements of the peripheral object were made in the order of decreasing visual angle.

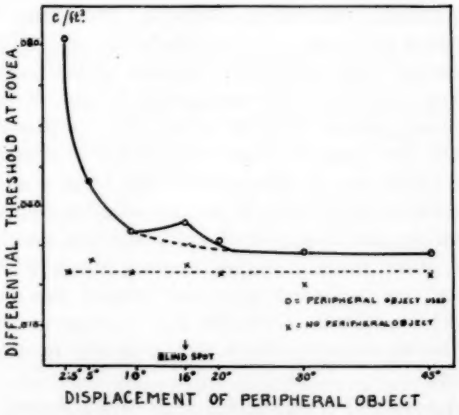

FIG. 4. The effect of displacing a bright object further and further toward the periphery upon the amount of stray light falling at the fovea, as measured by the change in the differential threshold. The amount of displacement is expressed in terms of the size of the visual angle subtended by the center of the test-object and the center of the peripheral object.

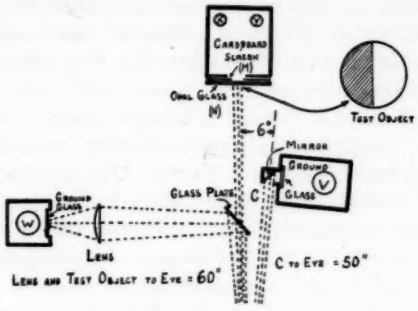

FIG. 5. Diagram of the apparatus for demonstrating nervous interaction of the periphery with the fovea as manifested by the effect of a bright peripheral object upon the differential threshold at the fovea when a bipartite test-object is used. The illuminated aperture, *C*, serves as the bright peripheral object. The test-object consists of a bright disk one-half of which is covered by rice paper. Light from source *W* serves as a substitute for stray light and makes it possible to determine the rôle played by stray light.

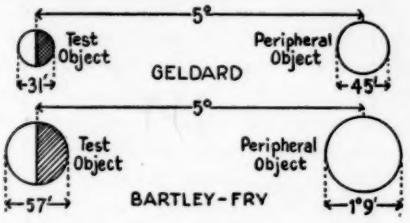

FIG. 6. A comparison between the conditions used by Geldard and the similar experiment by Bartley and Fry. In both cases the dimmer half of the test object was 0.59 as bright as the other.

The results show that the intensity of stray light falls off rapidly 5° to 10° away from the test object, and from there on shows only a slight decrease. It can be noticed by the observer that the image of the bright peripheral object is surrounded by a bright halo which diminishes rather rapidly between 3½° and 5°. At 16° the image falls on the blind spot. In an ophthalmoscopic examination of the eye the optic disk appears brighter than its surroundings and it might be expected that the amount of light reflected from the optic disk might be greater than other parts of the retina. The rise in the threshold actually found at 16° may be of significance in this connection.

So far we have not considered the possibility that the bright light in the periphery might affect vision at the fovea on account of nervous interaction of the peripheral part of the retina with the fovea. Geldard[6] has demonstrated a phenomenon which seems to indicate that such interaction might affect the threshold at the fovea when a bipartite test-object is used.

In order to duplicate the conditions used by Geldard, we set up the apparatus illustrated in Fig. 5. The test-object was provided by a circular aperture (1 in. in diameter) in the cardboard screen *M* which allowed a disk-shaped area of the milk glass *N* to receive illumination from the light sources, *X* and *Y*, the intensity of which could be varied by rheostat by the observer. Rice paper covered one-half of the test-object so that this half was only 59 percent as bright as the other. A point at the center of the test-object was fixated. The problem for the observer consisted in adjusting the illumination of the test-object till it appeared just noticeably divided into two parts. A bright object in the periphery

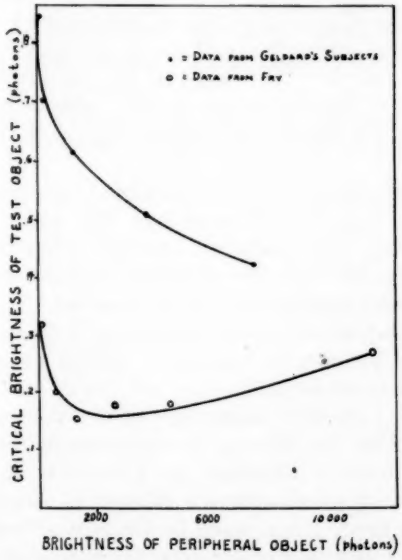

FIG. 7. A comparison of the results obtained by the present investigators with those obtained by Geldard in testing the effect of varying the brightness of a peripheral object upon the differential threshold at the fovea as measured with a bipartite test-object. The effect upon the differential threshold is expressed in terms of the brightness of the brighter half of the test object at which the two halves become just distinguishable, when the brightness ratio of the two halves is kept constant at 0.59.

[6] F. A. Geldard, *Brightness Contrast and Heyman's Law*, J. Gen. Psych. 5, 191–206 (1931).

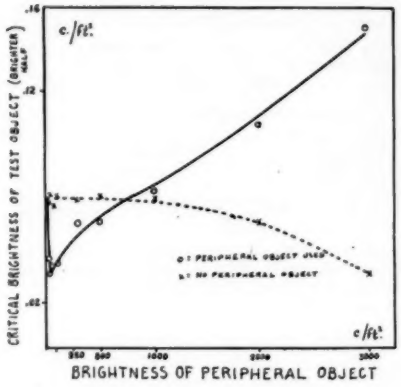

FIG. 8. The effect of varying the brightness of a peripheral object upon the differential threshold at the fovea, as measured with a bipartite test-object.

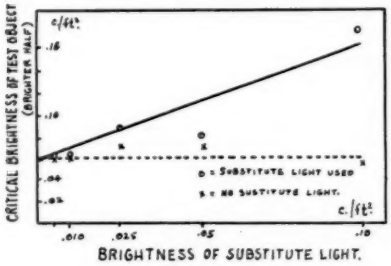

FIG. 9. The effect of varying the brightness of the "substitute stray light" upon the differential threshold at the fovea as measured with a bipartite test-object.

was provided by the illuminated aperture *C* which was 1 in. in diameter. The observer's head was held steady in a headrest. The left eye was shielded and the right eye peeped through an artificial pupil 2.33 mm in diameter.

The apparatus we have used may be compared with Geldard's in regard to certain essential points as shown in Fig. 6.

Fig. 7 shows a comparison of the results obtained by the different investigators in testing the effects of changing the brightness of the peripheral object upon the differential threshold at the fovea as measured by the critical brightness of the test object (the brightness of the brighter half of the test-object at which the two halves are just distinguishable). The intensity values are expressed in photons[7] to make the two sets of data comparable. Although the absolute values do not correspond, both curves begin by falling. The curve for Fry shows a subsequent rise, and such a rise might have occurred in the case of Geldard's subjects if a larger range of brightnesses for the peripheral object had been used. The fact that the rise in Fry's curve comes earlier than in the case of Geldard's subjects is probably due to the fact that the area of Fry's peripheral object is larger.

We have used a more extended range of intensities for the peripheral object than Geldard. The effects obtained with our extended range are shown in Fig. 8. The curve for Fry in Fig. 7 represents only a small fraction of the curve in Fig. 8.

In order to ascertain whether stray light falling at the fovea can account for the curve in Fig. 8, we have investigated the effect of uniformly illuminating the region of the fovea with a known quantity of light. Light from the lens (see Fig. 5) is reflected by the plate glass to the eye and illuminates the region surrounding and including the fovea. Since this light is equivalent to stray light falling on the fovea, let us, for convenience,

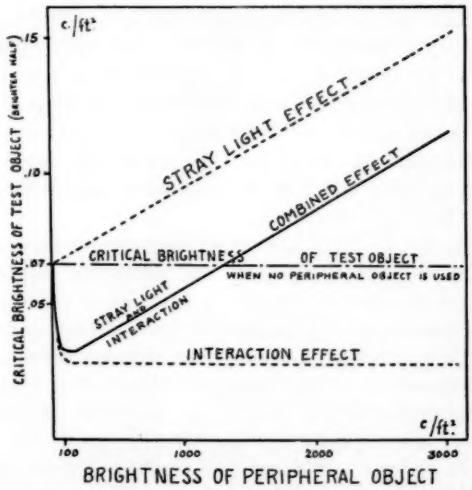

FIG. 10. An analysis of the curve in Fig. 7 showing the relative rôles played by stray light and nervous interaction.

[7] We have used the unit, photon, as defined by Troland as the retinal illumination produced by a surface having a brightness of 1 c per sq. m when the pupillary aperture is 1 sq. mm (L. F. Troland, *On the Measurement of Visual Stimulation Intensities*, J. Exper. Psych. 2, 1–33 (1917)).

call it "substitute" stray light. The only difference is that the bright stimulus in the periphery, which might produce interaction with the fovea, is absent. The effect of varying the intensity of the "substitute" stray light was tested. The results are shown in Fig. 9. They show a gradual increase in the critical brightness of the test object without any trace of an initial decrease at the lower end of the scale, such as occurs when a bright peripheral object is used.

The curve in Fig. 8 might be analyzed as illustrated in Fig. 10 into two components representing stray light and retinal interaction. It might be supposed that the part of the retina receiving the image of the bright spot interacts with the fovea and enhances the ability of the fovea to establish a brightness-contrast-border.

We have found no indication of this retinal interaction effect in our experiments with the disk-annulus test object. This effect seems therefore to be tied up with the use of the bipartite test-object. What constitutes the essential difference between the two kinds of test-objects merits further consideration.

Summary

In order to measure the intraocular stray light a bright object was placed in the periphery of the field of vision and the intensity of the stray light falling on the fovea was determined by measuring the change in the differential threshold. The change in threshold produced in this way can be compared with the change occurring when the region of the fovea is uniformly stimulated with a known intensity of light. It has been calculated from our results that a bright object subtending a solid visual angle of 1 millisteradian and having a brightness of 1 c per sq. ft. is capable of producing 0.00408 photons of stray light at a point on the retina 5° away from the image of the bright object, when the beam of light entering the eye is 1 mm in diameter.

The distribution of the stray light was also measured. The amount of stray light rapidly diminishes out to 10° and from there on diminishes only slightly. This corresponds to the subjective appearance of stray light, for the whole field is seen to be dimly illuminated with a bright halo immediately surrounding the bright object.

The possibility that the periphery affects the fovea by means of nervous interaction has been considered. Although this factor comes into play when a bipartite test object is used, it seems to be absent in the case of the disk-annulus test-object which we have used.

The writers wish to thank Professor P. W. Cobb for his helpful suggestions.

An Auxiliary Lens for Use with the Concave Grating

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(Received September 24, 1934)

A discussion of the elementary theory of the astigmatism of the concave grating. A sphero-cylindrical lens placed in front of the plate serves to produce a stigmatic image of the slit and increases the speed of the instrument by about 25× and doubles the dispersion, allowing the full theoretical resolving power to be realized.

I. INTRODUCTION

THERE are always two big problems facing the user of the concave grating; namely, the low intensity ordinarily obtained because of the small aperture of the instrument and its astigmatism; the other, the realization of the theoretical resolving power of the instrument. The first necessitates long exposures when one is dealing with weak sources, and consequently requires accurate temperature control of the grating in order to insure sharpness of lines. The second is made difficult by the lack of a photographic material with as high a resolving power as the grating itself.

Several suggestions have been made to overcome these two difficulties. Runge and Mannkopff[1] have discussed the problem in detail and have used a spherocylindrical lens between the source and slit, at such a position that a stigmatic image is produced by the grating. Lacroûte[2] has suggested the use of a cylindrical convex lens between the slit and grating, casting an image of the source on the slit with a spherical lens, and thus counterbalancing the astigmatism produced by the grating. Both of these methods give stigmatic images over only a short range of wave-lengths because of the chromatic aberration of the lens. Oldenberg[3] uses a convex cylindrical lens between the grating and plate. Lacroûte's method, however, introduces the difficulty that when the grating has been focussed in the usual way the focal curve is displaced by the lens, due to the extra length of optical path introduced by the lens. Any of these methods give a greatly increased intensity of image.

In order to realize the theoretical resolving power of the grating, Back[4] has made use of a concave lens between the grating and the photographic plate, producing a magnified image of the line beyond the usual position of the plate. But here we have introduced the disadvantage that the intensity of the lines is actually decreased, and the method is applicable to only a very narrow range of wave-lengths.

For some time we have been using a combination of the ideas involved in Back's and Oldenberg's methods, giving increased dispersion on the photographic plate and at the same time eliminating the astigmatism so that exposures are reduced to a small fraction of the usual time. This method consists in using a sphero-cylindrical lens between the grating and the plate and adjusting the lens so that a more convergent beam is produced vertically and a more divergent beam horizontally, the two foci coming at the same place. Before discussing the design of this lens, let us first consider the elementary theory of the concave grating.

II. THEORY OF THE CONCAVE GRATING

(a) The focal curves of a concave grating

Using the symbols θ and φ for the angles of incidence and diffraction, and s and t for slit and focal distances, we find that wave-lengths λ placed at a distance s from the grating will be brought to a focus in the principal plane of the grating on a curve given by the relation (to second order terms)

$$\frac{\cos^2\theta}{s}-\frac{\cos\theta}{\rho}+\frac{\cos^2\varphi}{t}-\frac{\cos\varphi}{\rho}=0. \qquad (1)^*$$

This represents what might be called the vertical

[1] Runge and Mannknopf, Zeits. f. Physik **45**, 13 (1927).
[2] Lacroûte, Comptes rendus **194**, 1803 (1932).
[3] Oldenberg, J. O. S. A. **22**, 441 (1932).

* See e.g., Baly, *Spectroscopy*, Vol. I.
[4] Back and Goudsmit, Zeits. f. Physik **47**, footnote p. 179 (1928).

focus of the grating; a point source at the slit becomes a vertical line at distance t. (ρ = radius of curvature.)

But at another point along the ray in the φ direction, at a distance R, there is a horizontal focus; that is, a point source at the slit becomes a horizontal line at distance R.

By the same method as used in deriving Eq. (1), it may be shown that the locus of the points R for different wave-lengths is given by (to second order terms)

$$\frac{1}{s}-\frac{\cos\theta}{\rho}+\frac{1}{R}-\frac{\cos\varphi}{\rho}=0. \qquad (2)$$

This calculation is easily carried out by considering that all rays striking the central line of the grating must have the same path length to the point R.

(b) The astigmatism of the grating

(I) With the help of Eqs. (1) and (2) the astigmatism of the grating for various types of mountings is immediately evident. From (1) we see that if $s=\rho\cos\theta$ then $t=\rho\cos\varphi$; that is, the principal focal curve is a circle of half the radius of curvature of the grating, called the Rowland circle. There are three types of mounting in which this property is used. Eq. (2) then becomes

$$\sec\theta-\cos\theta=\cos\varphi-\rho/R \qquad (3)$$

or

$$R=\rho/(\cos\varphi-\sin\theta\tan\theta), \qquad (4)$$

which is the equation of an hyperbola which has its minimum R at $\varphi=0$; that is, normal to the grating

$$R_{\min}=\rho/(1-\sin\theta\tan\theta) \qquad (5)$$

and is symmetrical with respect to the normal.

(*1*) *The Rowland mounting.* In this mounting $\varphi=0$ and θ varies. If the length of the ruling is h, then a plate set on the Rowland circle a distance ρ from the grating will intercept a beam in the vertical direction whose length is

$$(R-\rho)h/R=h\sin\theta\tan\theta \qquad (6)$$

the astigmatism of the Rowland mounting.

(*2*) *The Eagle mounting.* In this mounting $\theta=\varphi$ and θ varies, and a plate set on the Rowland circle at a distance $\rho\cos\theta$ from the grating intercepts a beam in the vertical direction whose length is

$$(R-\rho\cos\theta)h/R=2h\sin^2\theta \qquad (7)$$

the astigmatism of the Eagle mounting.

(*3*) *The Paschen-Runge mounting.* In this mounting the slit and plate holders are set permanently on the Rowland circle and the plate at $\rho\cos\varphi$ intercepts a beam whose length is

$$(R-\rho\cos\varphi)h/R=h(1-\cos^2\varphi+\sin\theta\tan\theta\cos\varphi), \qquad (8)$$

the astigmatism of the Paschen-Runge mounting.

(II) Meggers has described a mounting of the grating in parallel light; that is, $S=\infty$ and the diffracted rays are observed normal to the grating; that is, $\varphi=0$, then (1) becomes

$$-\cos\varphi/\rho+1/t-1/\rho=0 \quad \text{or} \quad t=\rho/(1+\cos\theta), \qquad (9)$$

the equation of a parabola with the minimum focal distance when $\theta=0$, $t=\rho/2$; that is, half the usual focal length of the grating. Under these conditions (3) becomes

$$-\frac{\cos\varphi}{\rho}+\frac{1}{R}-\frac{1}{\rho}=0, \quad R=\frac{\rho}{1+\cos\theta} \qquad (10)$$

and R and t are at the same place; that is, for the wave-length normal to the grating, the instrument is stigmatic.

(c) The auxiliary lens

The grating at the Mendenhall Laboratory is set in a Paschen-Runge mounting with an angle of incidence of about 48°, so that (4) becomes at $\varphi=0$

$$R_{\min}=\rho/(1-0.743\times1.11)=5.52\rho, \qquad (11)$$

which means $R_{\min}$ for a 21-foot grating is about 115 feet, or 94 feet behind the principal focal curve. For any short focal length lens this may

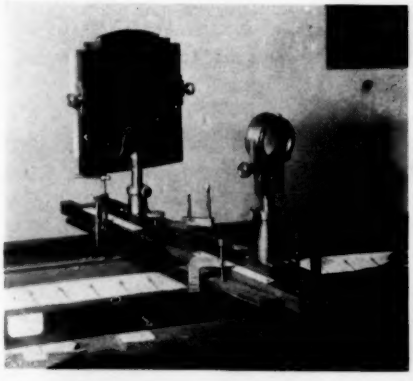

FIG. 1. The lens bed clamped to the grating table.

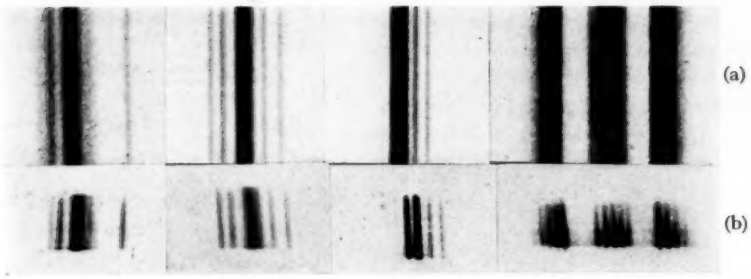

FIG. 2. FIG. 3. FIG. 4. FIG. 5.

FIG. 2. (a) Hg 5461 II order without lens. (b) Hg 5461 II order with lens. The increase in resolving power is evident.

FIG. 3. (a) Hg 4358 II order without lens. (b) Hg 4358 II order with lens. The faint satellite of the strong central component appears as a shading without the lens, but is clearly a separate line with the lens.

FIG. 4. (a) Hg 4047 II order without lens. (b) Hg 4047 II order with lens. The spaces between the lines are much clearer with lens.

FIG. 5. (a) Cd 3610 II order Zeeman effect at about 37,000 gauss. Without lens. (b) The same line using the lens. The improvement is obvious.

be considered at an infinite distance so that practically parallel light in a vertical plane is coming to a horizontal focus on the principal focal curve.

By means of the auxiliary lens we wished to increase our dispersion by a factor of two and also have a stigmatic image of the slit. The lens must therefore be negative for the horizontal beam and positive for the vertical beam. The combination finally chosen had a power of -3 diopters axis vertical and $+3$ diopters axis horizontal.

This lens was mounted in an optical bench type of mounting, as shown in Fig. 1. This allowed an adjustment of the lens along the line of sight, a vertical adjustment, and a rotation adjustment about its axis. A $3\frac{1}{4}$ inch by $4\frac{1}{4}$ inch plateholder was mounted in a suitable stand with elevating and sliding adjustments.

In adjusting the lens the following procedure was followed. First the bench bed was lined up exactly with the line of sight and two 50 cm scales were clamped one on each side so that the bench could slide as a whole toward or away from the grating. Secondly, the lens was clamped to the bench and set with its cylindrical axes perpendicular and horizontal. The focal lengths of our lens being about ± 33 cm, it was placed at about half this distance ahead of the focal curve of the grating. The third adjustment consisted of finding the stigmatic focal distance between the lens and the plate. This was quite critical as the beam intercepted was about 4.0 cm wide vertically and the lens had a relatively short focal length. This adjustment was best made photographically. Once this stigmatic separation was found the whole lens bench was moved in steps of 2 mm and a series of photographs of the spectrum line made. This last adjustment was not particularly critical. Finally, the lens was rotated until the sharpest image was obtained.

Figs. 2 ab, 3 ab, 4 ab and 5 ab show samples of the results obtained.

The increase in speed obtained is about a factor of 20 to 30. The increase in resolution is about the same as that obtained by increasing the order of the spectrum to the next higher order.

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